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**Nuclear Science Series  
Report Number 29**

# **Penetration of Charged Particles in Matter**

D652  
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1958

**National Academy of Sciences—  
National Research Council**

Publication 752

AUG 12 1960

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Nuclear Science Series

Report Number 29

*National research Council*  
Subcommittee on Penetration  
of Charged Particles in Matter  
Committee on Nuclear Science

# Penetration of Charged Particles in Matter

Proceedings of an Informal Conference  
Gatlinburg, Tennessee, September 15-18, 1958.

Edwin A. Uehling, Editor //

Publication 752  
National Academy of Sciences—National Research Council  
Washington, D. C.  
1960



Library of Congress Card Catalog Number 60-60021



D652  
N292  
1958

## FOREWORD

The Gatlinburg Conference of September, 1958, brought together about fifty people from Canada, Europe, Asia, and the United States who are presently active in the investigation of current problems in the penetration of charged particles in matter. About twenty-five scheduled talks on topics ranging from the reliability of experimental data to new approaches in the theoretical description of energy loss mechanisms were presented. In addition there were unscheduled talks and informal discussions during free intervals of the four-day period.

The conference was organized by the Subcommittee on Penetration of Charged Particles in Matter of the National Academy of Sciences--National Research Council, and was sponsored by the Atomic Energy Commission, National Science Foundation, and the Air Force Office of Scientific Research.

The need for this conference had been felt for some time and tentative suggestions in regard to it had been made by a number of people during the last decade. One of the factors responsible for these suggestions was the growing conviction that a new synthesis of experimental and theoretical information was possible. On the one hand the experimental data were becoming more accurate and reliable. A deliberate effort was being made to compare data obtained in different laboratories, and, where discrepancies existed, vigorous efforts had been made to determine the causes which were responsible. Thus new information of greatly increased accuracy was constantly becoming available. Simultaneously with these developments the ability to make use of the experimental information which was available had increased. The magnitude and the method of application of the various kinds of corrections which intervene between the idealized theory on the one hand and the experimental results on the other had been studied extensively in recent years and the consequences of applying these corrections to good experimental data were just beginning to be felt. The hope of speeding up this process which would lead eventually to the determination of the numerical values of important parameters in the energy loss relations was one of the primary objectives of the conference.

There had also been developments along other lines which it was felt could be profitably reviewed. On the experimental side these included such diverse topics as the determination of capture and loss cross sections, the measurement of the energy required to produce an ion pair, and the energy loss of a charged particle in a single collision. On the theoretical side there had been advances in the study of transitions induced in simple atomic systems by low energy radiations, and there had been advances also in the study of very complex systems in which the attempt is made to follow various details in the energy degradation problem. It is clear that the objectives of the conference ranged over quite a wide front.

The outline of topics discussed is given in the following table of contents. The written reports on these topics have been prepared by the speakers or by the chairmen of the various sessions in collaboration with the speakers. In some instances the reports include the results of subsequent discussions. In a few other cases the reports have been shortened or reduced to an abstract because of subsequent adequate publication elsewhere. References to these publications are of course included.

Various members of the subcommittee acted as the organizers and chairmen of the various sessions and assisted in the preparation of this report. Over-all responsibility for the report as presented rests, however, with the chairman.

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Committee on Nuclear Science

Edwin A. Uehling, Chairman  
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## I. STOPPING POWER AND RANGES

### 1. Stopping Power at Low Energies

Ward Whaling  
California Institute of Technology

The experimental methods used in recent measurements of low energy stopping power will be described and the results obtained by these methods will be discussed. Particular attention will be given to the consideration of questions of reliability. We will find that there is a considerable body of compatible data obtained by various experimenters in different laboratories. We will also find instances in which good agreement does not exist and in which the sources of the discrepancies are still unknown. Some questions of interpretation and the need for further experimental information will be mentioned.

Measurement of the stopping cross section  $1/N(-dE/dR)$  requires the determination of two quantities; namely,  $\Delta E$  and  $N \Delta R$ . Let us consider first the measurement of  $\Delta E$ . A typical experimental arrangement is that employed at the University of Chicago and illustrated in Figure 1.<sup>1</sup> The beam from a Van de Graaff or Cockcroft-Walton accelerator of known energy is scattered from a thin layer of gold to reduce the intensity of the beam. The energy spread introduced by the thin scattering foil is insignificant. One measures the spectrum of the particles scattered into the analyzer. The energy of this group of particles is shifted to a lower energy when the stopping foil is interposed and the energy shift is measured with the analyzer. In order to study the stopping power of gases, the foil is replaced by a differentially pumped cell without window foils, and the energy shift due to the gas in the cell is measured. A similar scheme is used by us at the California Institute of Technology and by Brolley

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<sup>1</sup>S. K. Allison and S. D. Warshaw, Revs. Mod. Phys. 25, 779 (1953). Fig. 1 is their Fig. 4, p. 788.



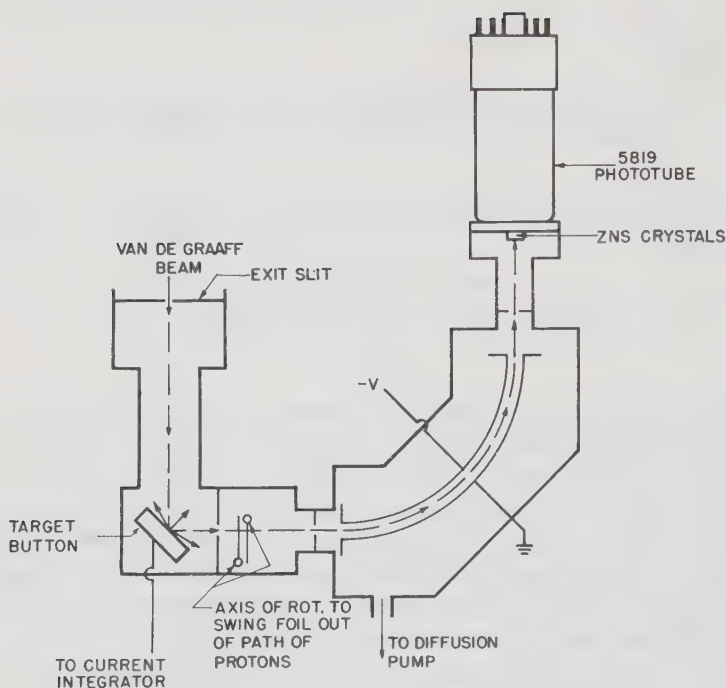


Figure 1. Typical experimental arrangement used at the University of Chicago for the measurement of  $\Delta E$ .

and Ribe at Los Alamos. In the CIT arrangement an electrostatic analyzer is used for the incident beam and a magnetic analyzer for the scattered beam. Brolley and Ribe use a No. 1 scintillation spectrometer to measure the energy loss when a gas cell is introduced.

A different scheme is used at Copenhagen and Ohio State. The incident beam of protons of known and variable energy is the same, but the target is one which exhibits narrow resonances for the  $(p, \gamma)$  reaction. A gamma detector enables one to determine accurately when the energy of the beam actually striking the target is at the known resonance energy. A foil is placed in the beam and the energy of the accelerator is increased to bring the energy of the beam striking the target back to the resonance energy. At Ohio State this method has also been used to measure the energy loss in gases by covering the opening to the target chamber with a gas tight foil and measuring the energy shift in the same way as before after introducing a gas.

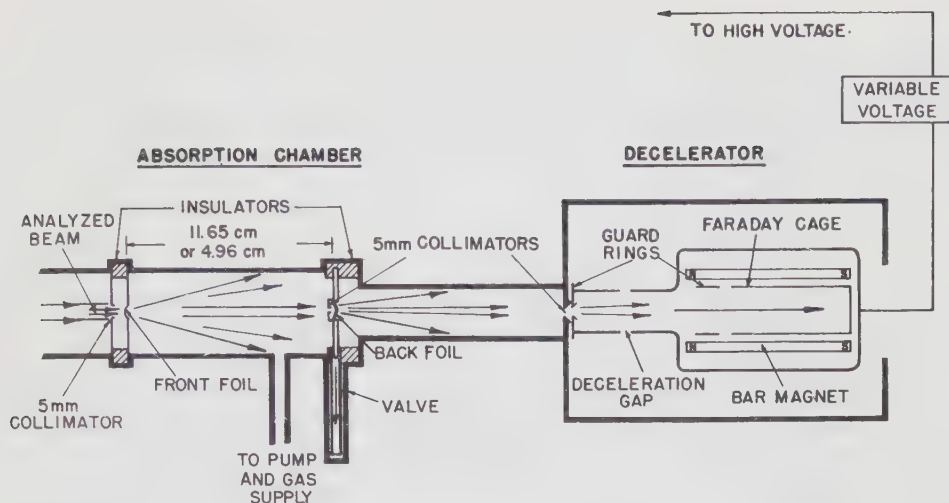


Figure 2. Typical experimental arrangement used at the Los Alamos National Research Laboratory for the measurement of  $\Delta E$ .

Phillips at Los Alamos has used a different scheme. His equipment is illustrated in Figure 2.<sup>2</sup> Protons or other hydrogen ions are accelerated through a potential drop to ground. They pass through a gas cell with windows and then are decelerated as they coast to a beam collector which is at the same high potential as that from which the ions started. If there were no energy loss in the cell, the ions would reach the collector as they stopped. If they lose energy in the cell the potential of the collector must be lowered in order to collect the beam. It is this change in collector potential which is measured. With this scheme Phillips can measure values of  $\Delta E$  of perhaps 1 kev to 25 ev.

With the exception of the Phillips scheme these methods are capable of measuring  $\Delta E$  with an accuracy of 1 to 3 per cent, depending on the magnitude of  $\Delta E$ . In order to insure a nearly true differential measurement one tries to make  $\Delta E$  small.

We turn now to the measurement of  $N \Delta R$ , the thickness of the stopping layer. This measurement is also made in a variety

<sup>2</sup>S. K. Allison and S. D. Warshaw, *Revs. Mod. Phys.* 25, 779 (1953). Fig. 2 is their Fig. 13, p. 799.

of ways. For gases it involves a determination of cell length, gas temperature and pressure. Sources of error include the incomplete determination of window bulge, gas impurities, and gas absorption on the windows. There are probably unknown sources of error. However, an accuracy of 1 per cent appears to be possible.

In order to determine  $N\Delta R$  for solids one weighs, or measures by chemical analysis, the amount of material deposited on a known area. Deposition is necessary because the stopping layers are usually too thin to be self supporting. Details of the method used at CIT can be illustrated by reference to Figure 1. A supporting foil is hung from a microbalance in the vacuum system. A large baffle with an opening of known area is placed in front of the foil and the stopping material is evaporated from a furnace on to the foil. The energy loss and weight can thus be measured without exposing the layer to air. Some of the evaporated material is also collected on the back of the scattering target. A nice feature of the method is that an energy analysis of the protons scattered from this layer makes possible an analysis of the composition of the material being evaporated. This analysis of composition is very comforting when one is working with a stopping material as active as lithium. However, the over-all reliability of these measurements for solids is probably not so high as for gases. The accuracy of measurement of the thickness of thin solid layers is probably no greater than 2 to 5 per cent.

There is another method of stopping power measurement which we have used at CIT which avoids the necessity for a determination of  $N\Delta R$ . According to this method one measures the ratio of the stopping cross section to a reaction cross section. Thus it is not an absolute measurement. The method appears to be a rather good one if the reaction used is a Rutherford scattering. There are certain problems connected with the use of this method, but it is surprising that it has not been used more extensively.

We turn now to a discussion of the results obtained by the various methods. Figure 3 shows the results from Chicago, Ohio State, Los Alamos, and CIT for protons in air. The over-all accuracy claimed by the various experimenters is 2 to 5 per cent. The internal consistency of the data suggests that this estimate of errors is a cautious one. Another test of the accuracy



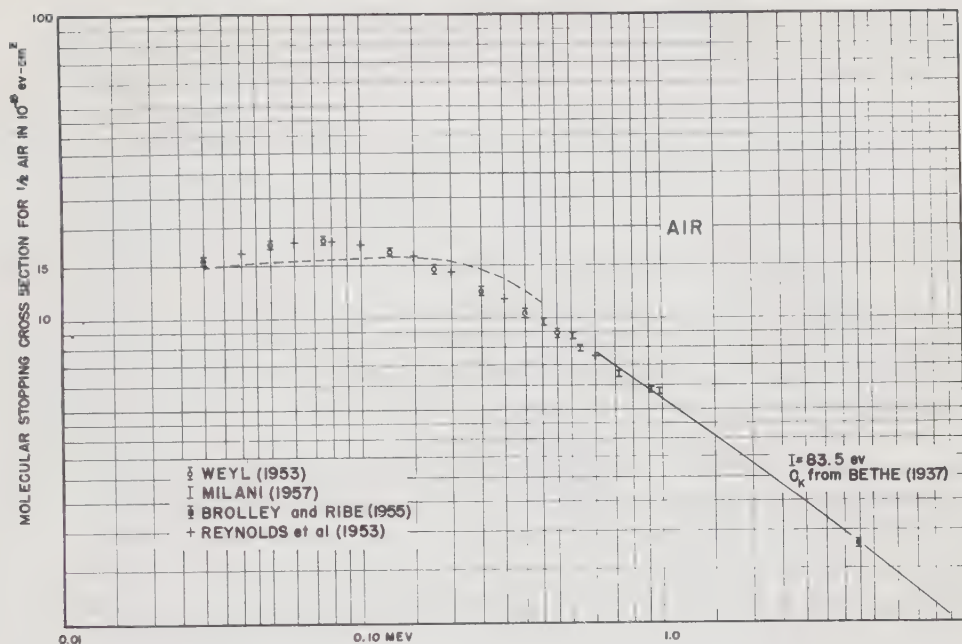


Figure 3. Stopping cross sections in air.

of the experimental data is obtained by integrating it to obtain a range energy curve. This curve agrees with the values obtained by Jesse and Sadauskis in 1950 within their error of 20 kev. The comparison with published theoretical curves is, however, not so satisfactory. If we restrict ourselves to the solid line curve obtained by Bethe in 1937 the agreement is good. The theoretical curve can in fact be used to span the gap in measurements between 1 Mev and 4 Mev. But the situation at lower energies is different. The dotted curve reproduces the theoretical relation published by Bethe in his 1949 Brookhaven report. This relation was obtained by differentiation of what was believed to be the correct range energy relation for protons in air, a relation which was itself based on a careful study of internal consistencies of theoretical and experimental information. However, subsequent information as obtained in the experiments described in Figure 3 has shown how inaccurate such relations can be. This is the region in which electron capture and loss and other effects become important and the theory is simply incapable of taking these effects into account with sufficient accuracy to provide a satisfactory comparison with the experimental data.

There are perhaps a dozen gaseous stopping materials for which the stopping cross sections are known with the same accuracy that has been indicated for air. Indeed, the cross sections are known over an even wider range of energy as a consequence of the low energy work of Phillips at Los Alamos. Figure 4 shows the results for hydrogen and helium. The low energy results of Phillips overlap nicely with the results obtained at CIT, and the latter in turn join smoothly to the theoretical curve established by the high energy point of Brolley and Ribe. However, the results for other gases are not so satisfactory. At very low energies the Los Alamos results lie as much as ten per cent below the CIT results. Only one gas has been measured at these low energies elsewhere. This is argon shown in Figure 5 which includes the data of Weyl at Chicago as well as values computed from a range table by Jorgensen in addition to the Los Alamos and CIT results. The Chicago measurements seem to support the CIT values. The values attributed to Jorgensen tend to be high, as he has pointed out, because his measurements reflect stopping effects not included in the other measurements. Further independent experiments at low energies would be very desirable.

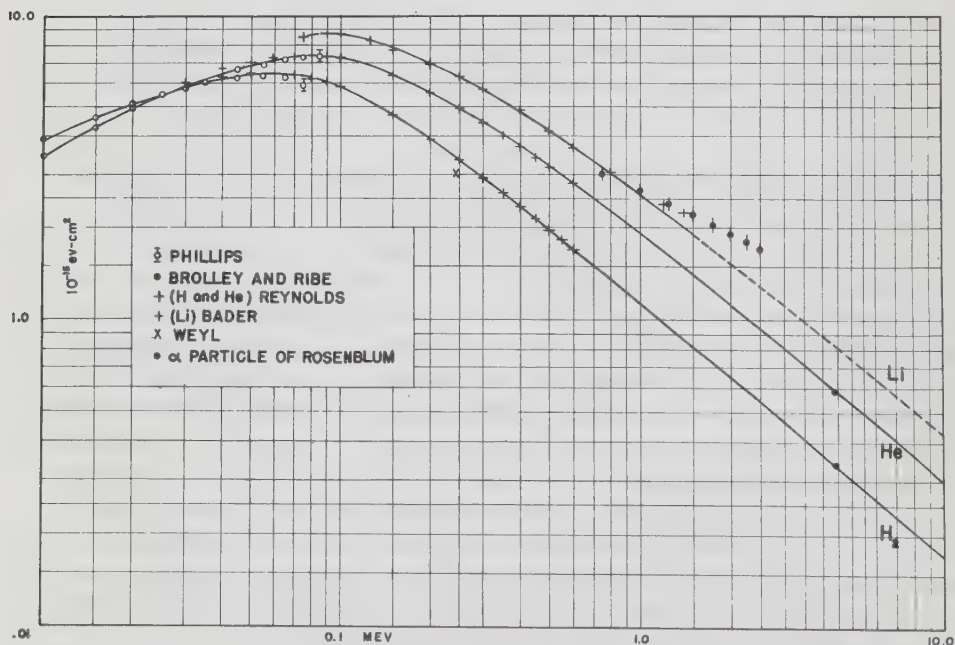


Figure 4. Stopping cross sections in  $\text{H}_2$ , He and Li.

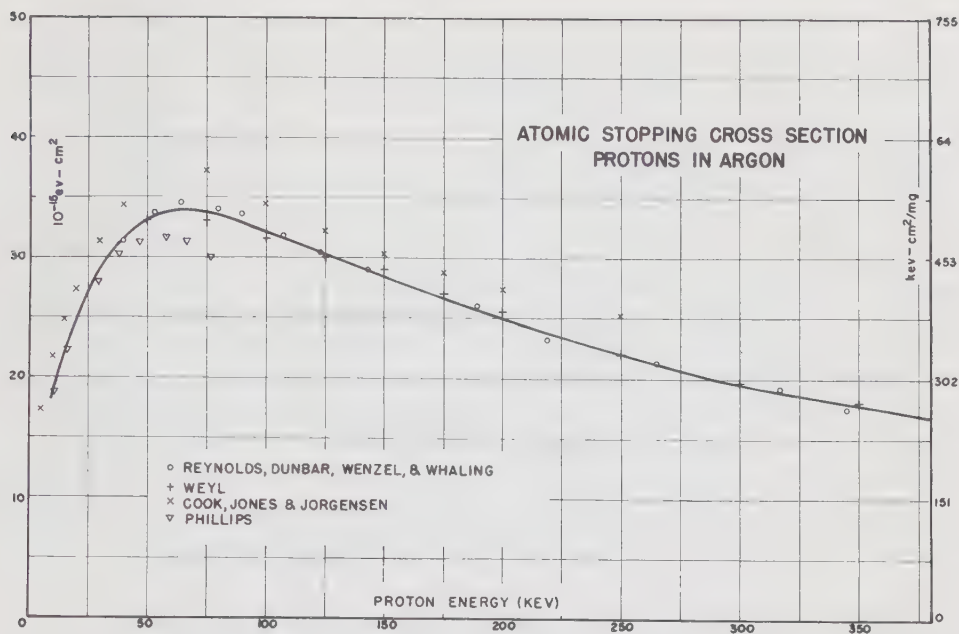


Figure 5. Stopping cross sections in A.

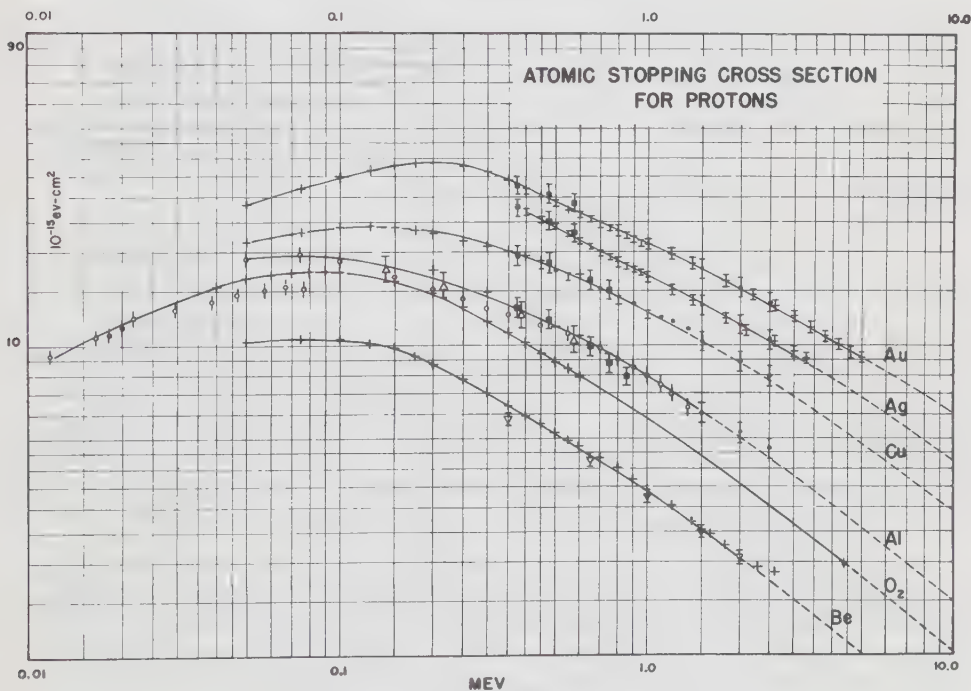


Figure 6. Stopping cross sections in metals.



In solid stopping materials, the experiments are more difficult and the results are not as accurate. Results for a number of metals are shown in Figure 6. When plotted in this way the results seem to be quite satisfactory with only minor discrepancies being exhibited. There are nevertheless wide variations among the results obtained in different laboratories. The worst case is that of gold. Figure 7 illustrates the present situation for this material. There are measurements from Copenhagen, Chicago, and Ohio State as well as CIT. All of the measurements are competently made, and the reasons for the discrepancies are not known. Allison has suggested that beaten gold foils do not give the same results as evaporated foils. However, the CIT measurements include both kinds with good agreement. The solid curve has been drawn deliberately to represent these latter measurements. One also notices that in the case of gold the curve is somewhat odd in shape. The slight dip in the neighborhood of 0.6 Mev which one observes in Figure 6 may be real.

The scope of the stopping power data using protons is shown in Figure 8. All recent measurements and the more accurate of the earlier measurements are included in this figure. There are large gaps in the available data both with respect to atomic number, and with respect to energy range covered. But there is enough data to provide some check on the theoretical ideas, particularly among the light elements. Boron has not been measured but is now being done at CIT. Fluorine also is missing, but  $\text{LiF}$  and  $\text{CaF}_2$  are common targets and data on the compounds are available.

Stopping power data at low energies for fifteen different media including both gases and solids are summarized in Figure 9. One of the striking features of this presentation is that there is obviously no simple interpolation procedure that can serve as a guide in going from one stopping medium to another. At these low energies the effects of capture and loss of electrons and of binding energy are such as to mask the simple  $Z$  dependence. In the absence of adequate theoretical guides there is consequently no alternative to experimental measurement.

The discussion so far has been concerned primarily with protons. When we go to alpha particles or heavier ions the situation is very much less satisfactory. There are practically no measurements. Even for alpha particles the available data for energies less than 2 Mev is very meager. Allison's group has studied H, He, A and air with alpha particles of 100 to 400 kev

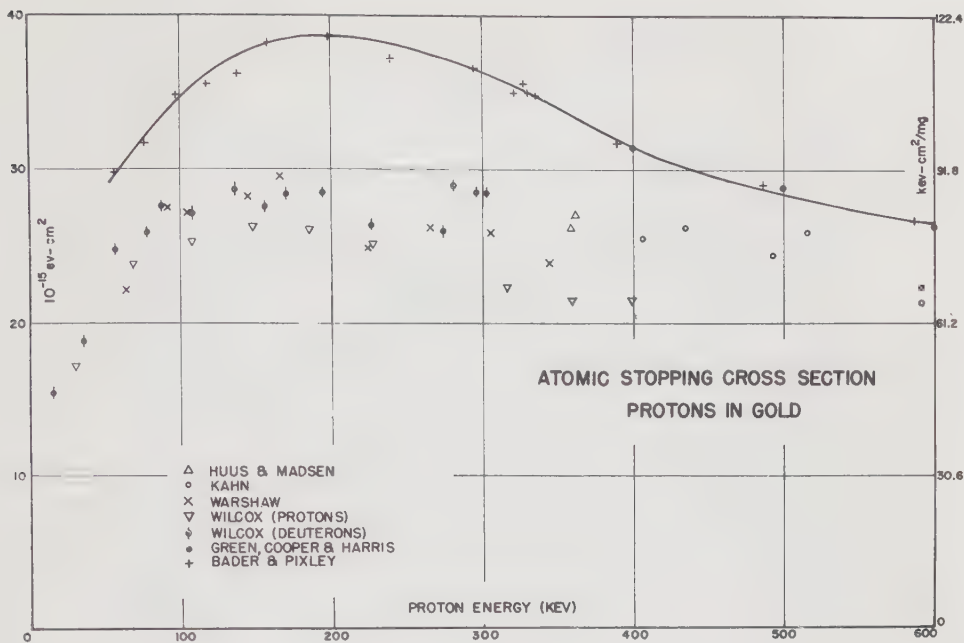


Figure 7. Stopping cross sections in Au.

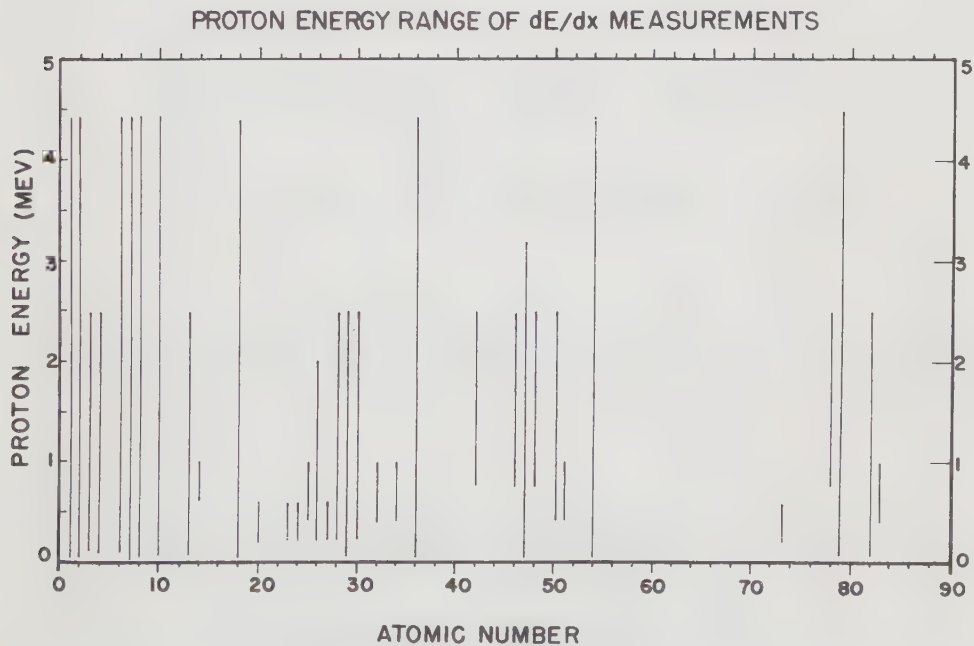


Figure 8. Scope of the available stopping power data for protons.

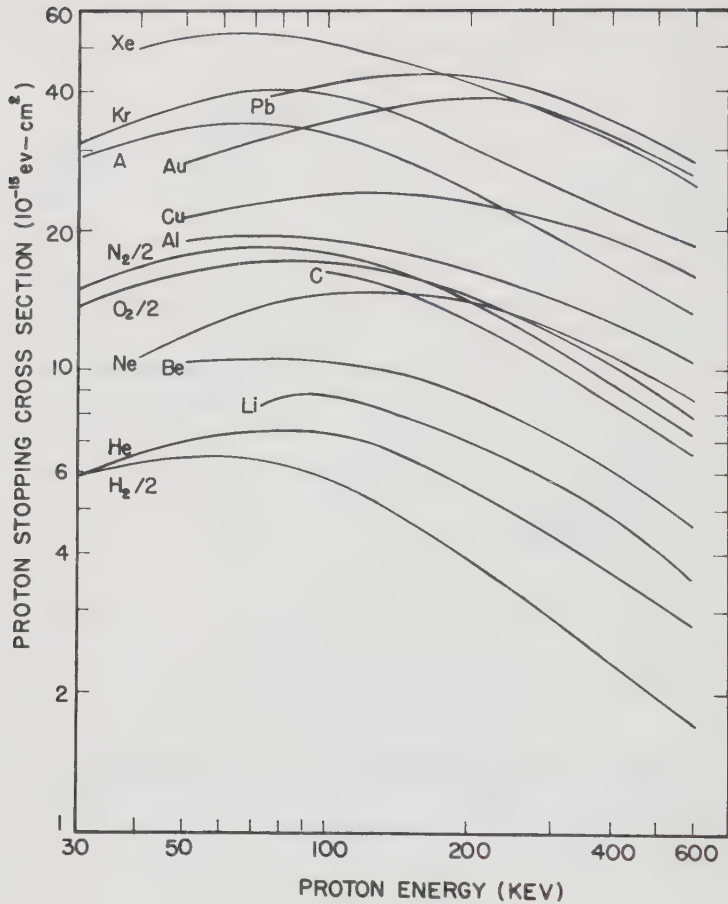


Figure 9. Stopping power data for fifteen different media including both gases and solids.

energy. Allison also has made measurements on these same four gases with  $\text{Li}^7$ ,  $\text{N}^{14}$  and  $\text{Ne}^{20}$  ions. There is very little other data at low energies. Also there is no theory that is valid in this range.

The absence of an adequate theory of stopping power at low energies makes further synthesis of the experimental results very difficult. However, one is tempted, even at these low energies, to use what information is available on K, L, and M shell binding energy corrections, but with neglect of capture and loss considerations, to extract from the data what information one can about the value of the mean ionization potential. The measurements of

Brolley and Ribe have been interpreted in this way by Walske with rather good results. We have also considered the data in the following way. Using the scheme of Lindhard and Scharff, we have plotted experimental values of  $EdE/NdR$  as a function of  $\log E$ . In the absence of binding energy and electron capture and loss effects the experimental points in such a plot would be expected to lie on a straight line. We then draw the best straight line through the points and read from the intercept  $E$  of the straight line with the horizontal axis the value of the mean ionization potential  $I$  as  $I = 4E(m/M)$ , where  $m/M$  is the ratio of electron mass to the mass of the incident particle. Some of the results obtained in this way are summarized in Figure 10. The variation of  $I$  with  $Z$  is about the same as has been observed before. There are of course stopping cross section measurements for many materials which are not included in Figure 10, and measurements made at energies which are so low that a straight line in the Lindhard-Scharff plot is not approximated have been excluded. A different kind of plot which is of some interest is the plot of stopping cross section as a function of  $Z$ . Such a plot for protons of 500 kev is shown in Figure 11. Since the energy is

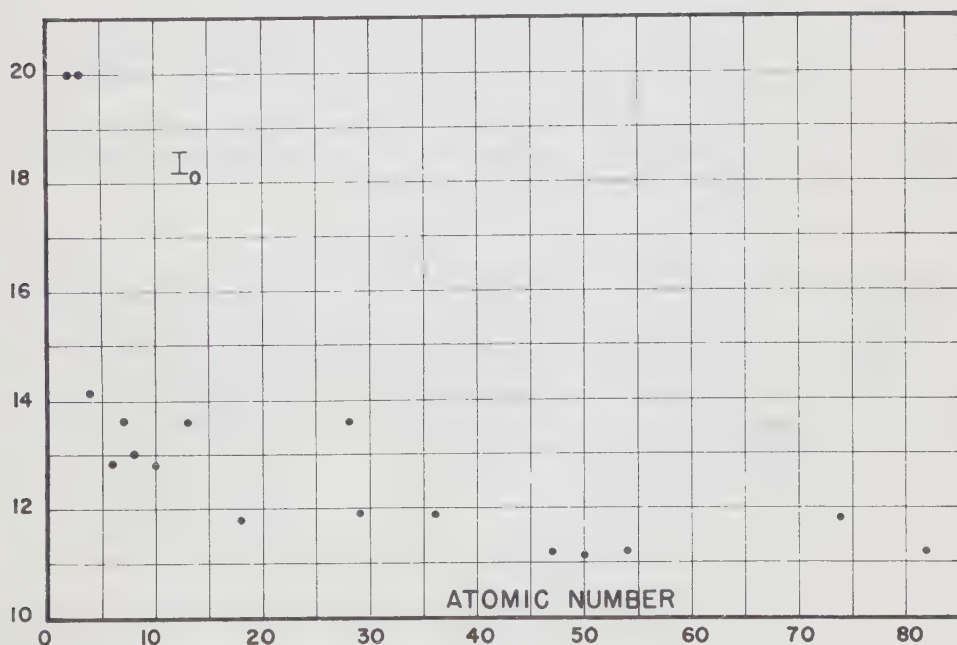


Figure 10. Experimental values of the Mean Ionization Potential as a function of Atomic Number and obtained by extrapolation from the Lindhard-Scharff plot.



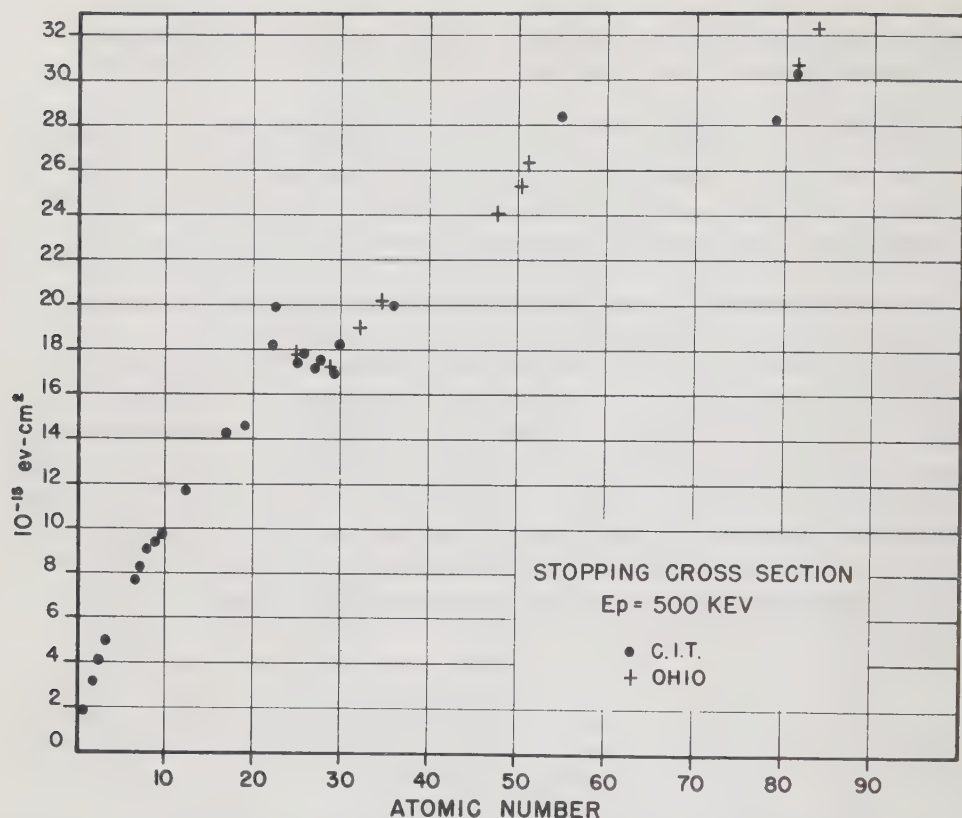


Figure 11. Atomic stopping cross sections for protons of 500 kev as a function of Atomic Number.

sufficiently high so that electron capture and loss effects are probably negligible, the discontinuities in the data are presumably due to the fact that the mean ionization potential  $I$  is not a monotonically varying function of  $Z$ . Cooper, who was then at Ohio State, was the first to observe these discontinuities which have been subsequently verified by us.

## 2. The Current Status of Direct Energy Loss Measurements in Pure Elements for Protons Above 5 Mev

K. R. MacKenzie  
University of California at Los Angeles

Accurate and reliable data on the rate of energy loss by heavy charged particles with an initial energy greater than 5 Mev are not very abundant. In recent years only six investigations<sup>3-8</sup> have been carried out in which the differential energy loss has been measured. Of these investigations, two were on an absolute basis; namely, the work of Brolley and Ribe on gases and the work of Sachs and Richardson on metals. The rest of the measurements were made relative to aluminum or copper.

Comparisons of the data obtained by different people and on different materials are most easily made if the data is plotted according to the scheme of Lindhard and Scharff.<sup>9</sup> A function proportional to the stopping power per electron, namely,  $L(x) = (mv^2/4\pi e^4 z^2) (1/NZ) (-dE/dR)$ , is plotted against the logarithm of a function  $x$  depending on energy and atomic number of the stopping material. If one assumes that the properties of the stopping atoms are adequately described by the Thomas-Fermi model

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<sup>3</sup>J. G. Teasdale, Office of Naval Research Report ONR-3 (unpublished).

<sup>4</sup>D. C. Sachs and J. R. Richardson, Phys. Rev. 83, 834 (1951) and 89, 1163 (1953).

<sup>5</sup>C. E. Bakker and E. Segré, Phys. Rev. 81, 489 (1951).

<sup>6</sup>C. P. Sonett and K. R. MacKenzie, Phys. Rev. 100, 734 (1955).

<sup>7</sup>J. E. Brolley and F. L. Ribe, Phys. Rev. 98, 1112 (1951).

<sup>8</sup>V. C. Burkig and K. R. MacKenzie, Phys. Rev. 106, 848 (1957).

<sup>9</sup>J. Lindhard and M. Scharff, Kgl. Danske Videnskab. Selskab Mat.-Fys. Medd. 27, No. 15 (1953).

one uses  $x = v^2 h^2 / e^4 Z$ . All of the available data can be included in such a plot. In the case of absolute measurements, one is making a direct plot of experimentally measured quantities multiplied by well-known constants. In the case of relative measurements one must choose the numerical value of  $L(x)$  for the standard substance at the energy of measurement. The theory of stopping power at medium energies including a knowledge of shell corrections and the presumed value of the mean ionization potential is used as a guide in making this normalization of the data. Finally, and in order to obtain further comparisons, data from range energy measurements which are not the main subject of this report may be incorporated into the plot. This also requires a kind of normalization. One procedure is to calculate the range from the stopping power relations in which the parameters are adjusted until the measured range is obtained. The stopping power relation, or rather  $L(x)$ , is then plotted.

Figure 12<sup>10</sup> shows the results using all of the data available at this time. Most of the measurements made subsequent to 1951 are displayed separately. These include in addition to the measurements reported in reference 3 to 8, some low energy measurements on copper and silver,<sup>11, 12</sup> and some previously unstressed measurements<sup>13</sup> made in 1949 using 37 Mev alpha particles. All relative data except that of Bakker and Segré have been normalized to  $I = 163$  ev for Al which is the value obtained in the absolute measurements of Sachs and Richardson<sup>4</sup> and is close also to the new value of  $I = 164$  ev obtained by Bichsel, Mozley and Aron in their range measurements.<sup>14</sup> The Bakker-Segré data were normalized to the range measurements of Mather and Segré;<sup>15</sup> i. e., to  $I$  for aluminum equal to 150 ev. If this data had been normalized instead to  $I = 163$  ev the points representing this data would have been lowered about 0.1 to a position a little closer to the straight line

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<sup>10</sup>Fig. 12 is Fig. 2 of Ref. 8.

<sup>11</sup>Chilton, Cooper, and Harris, Phys. Rev. 93, 413 (1954).

<sup>12</sup>Green, Cooper, and Harris, Phys. Rev. 98, 466 (1955).

<sup>13</sup>E. L. Kelly, Phys. Rev. 75, 1006 (1949).

<sup>14</sup>Bichsel, Mozley and Aron, Phys. Rev. 105, 1788 (1957).

<sup>15</sup>R. Mather and E. Segré, Phys. Rev. 84, 191 (1951).

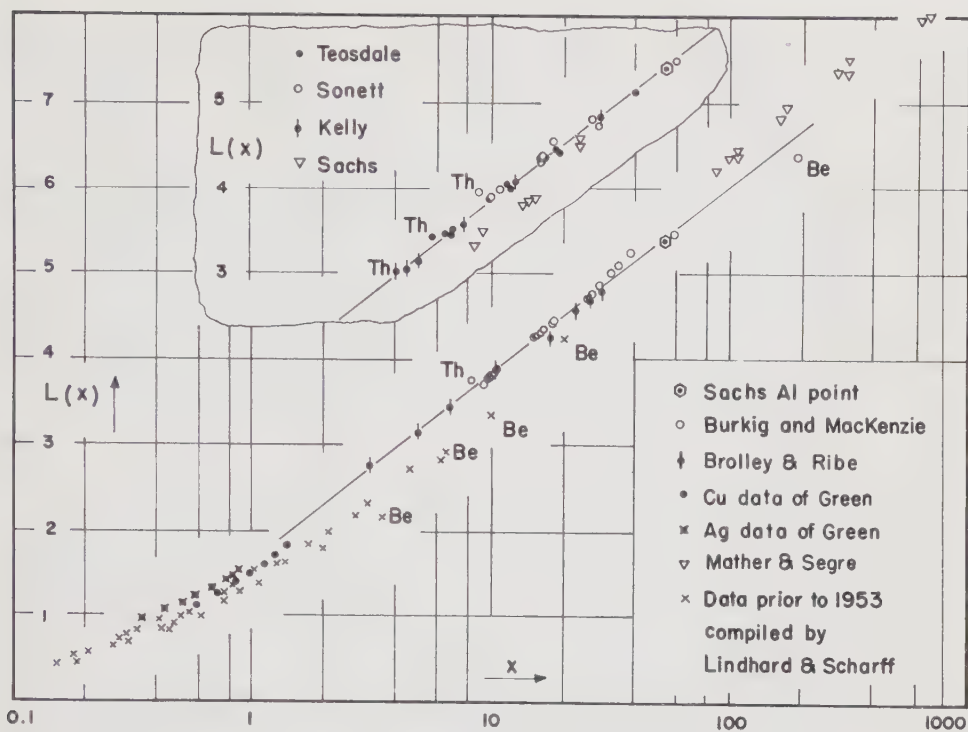


Figure 12. Lindhard-Scharff plot of the available data on energy loss.

drawn through the other points but not on it. In order to display individually all of the points in Figure 12 most of the data obtained before the most recent data of reference 8 has been displaced vertically.

One observes that the picture has changed considerably since 1953 when Lindhard and Scharff<sup>9</sup> proposed this procedure for examining the data. The crosses of Figure 12 represent the low and medium energy data available to these authors at that time. This data suggested the possibility of a universal curve in which the departure from a straight line relation in the Lindhard-Scharff plot would be quite considerable in the range of  $x$  in which binding energy effects are important. Recent data shows that the values of  $L(x)$  in the range of  $x$  from 1 to 50 lie higher than was believed to be the case before. One observes that this data joins smoothly with the low energy points of Green, Cooper, and Harris. However, it appears unlikely that a single curve can represent all of the data. While some of the deviations are likely to be due to errors and



unknown factors of measurements, other deviations are undoubtedly real. We can distinguish here between deviations such as the high energy Bakker - Segré data which we will not be able to interpret fully until more high energy data becomes available and the deviations between the heavy element data of Sachs and his own measurements on the light elements as well as other measurements made in the same energy range. Since all of the latter has been normalized to Sachs' aluminum point it is difficult to understand why his similar measurements on the heavy elements, Rh through Au, should give results which differ so much from the other data. Richardson (private communication) has pointed out that possibly a correction for multiple scattering should have been applied to the heavy element data.

We have made references throughout this discussion to normalization of the data which involve the assignment of a numerical value to the mean excitation potential  $I$  of a reference substance. We should now note specifically that the numerical values which we have mentioned are considerably higher than the values believed to be correct a few years ago. The evidence that the value of  $I$  for aluminum, for example, is higher than Wilson's value of 150 ev has been growing steadily since the range measurements of Bloembergen and van Heerden<sup>16</sup> and the stopping power measurements of Hubbard and MacKenzie<sup>17</sup> and of Sachs and Richardson suggested that the true value should lie 10 per cent or more higher. Subsequently, Caldwell<sup>18</sup> has analyzed the data and shown that all of the mean excitation potentials should be higher than was previously believed to be correct and that the new values would serve also to achieve a higher degree of consistency in the measurement of the masses of the K meson and other new particles. Finally, the accurate range measurements of Bichsel<sup>14</sup> confirm the higher range of the new values and tend to render their actual numerical determination more precise.

Most of the measurements, including even the high energy measurements such as those of Bakker and Segré, require an

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<sup>16</sup>N. Bloembergen and P. J. van Heerden, Phys. Rev. 83, 561 (1951).

<sup>17</sup>P. L. Hubbard and K. R. MacKenzie, Phys. Rev. 85, 107 (1952).

<sup>18</sup>D. O. Caldwell, Phys. Rev. 100, 291 (1955).

evaluation of binding energy corrections as an essential step in the determination of an energy independent mean ionization potential. Here too there have been important advances and changes in procedures in recent years. The theory of Walske<sup>19</sup> provides the numerical values of the K and L shell corrections and the basis for estimates of the corrections for higher shells. To an ever increasing extent these corrections are being used when experimental data are analyzed and the value of  $I$  is determined. The corrections are not large, being perhaps of the order of 0.1 in the vertical scale of Figure 12 at  $x = 20$  where  $L(x)$  lies between 4 and 5. The application of this correction consequently requires about a 10 per cent increase in  $I$  to preserve the measured  $L$  value. The data of Figure 12 as analyzed by Caldwell and others has had the binding energy corrections applied. An example of the application of such corrections is as follows. Measurements of stopping power are made relative to aluminum at, say, 20 Mev. With faith in the value of  $I$  for aluminum equal to 163 ev as determined from the absolute stopping power of Sachs or the range measurement of Bichsel, one assigns a value to  $L(x)$  for Al which is the difference of two terms, one depending on  $I$  and the other being the calculated binding energy correction. The point representing  $L(x)$  for the substance being measured is then the product of the  $L(x)$  for the standard substance as corrected for binding multiplied by the measured relative stopping power.

In spite of the increased accuracy of the more recent data, the more consistent application of binding energy corrections, and the independent evidence in regard to the value of the mean excitation potential, it is not yet possible to say that a unique value for each  $Z$  can be assigned. Rough calculations suggest that for  $Z > 13$ ,  $I/Z$  does not change much, or at least that there is no pronounced over-all trend. The values quoted for  $I/Z$  have been as large as 14, but the present tendency is toward a somewhat lower value (although higher than the value 11.5 used in the calculation of the range energy tables of Aron, Hoffman and Williams,<sup>20</sup> and certainly higher than the value of 9.5 which is obtained for the heavier elements if  $I$  for Al is taken to be 150 ev). Further discussions of the determination of  $I$  for the various elements and the various categories of stopping materials are likely to be given in the succeeding sessions.

<sup>19</sup>M. C. Walske, Phys. Rev. 88, 1283 (1952) and 101, 940 (1956).

<sup>20</sup>Aron, Hoffman, and Williams, Atomic Energy Commission Report AECU-663 (unpublished).

We turn now to one other aspect of the observations. The data of Burkig and MacKenzie<sup>8</sup> are believed to be of sufficient accuracy to exhibit meaningful fluctuations of the stopping power as a function of  $Z$  which may be superimposed on the systematic trend. When this data for the stopping power relative to Al is plotted on an expanded scale we obtain the curve shown in Figure 13.<sup>21</sup> The ordinate in this figure is  $\log Z$ . The Be point is omitted from this figure.

Individual fluctuations lying outside of the experimental error are now easily detected. The most striking variation occurs in the region of the transition elements from Ca to Ni. There is a

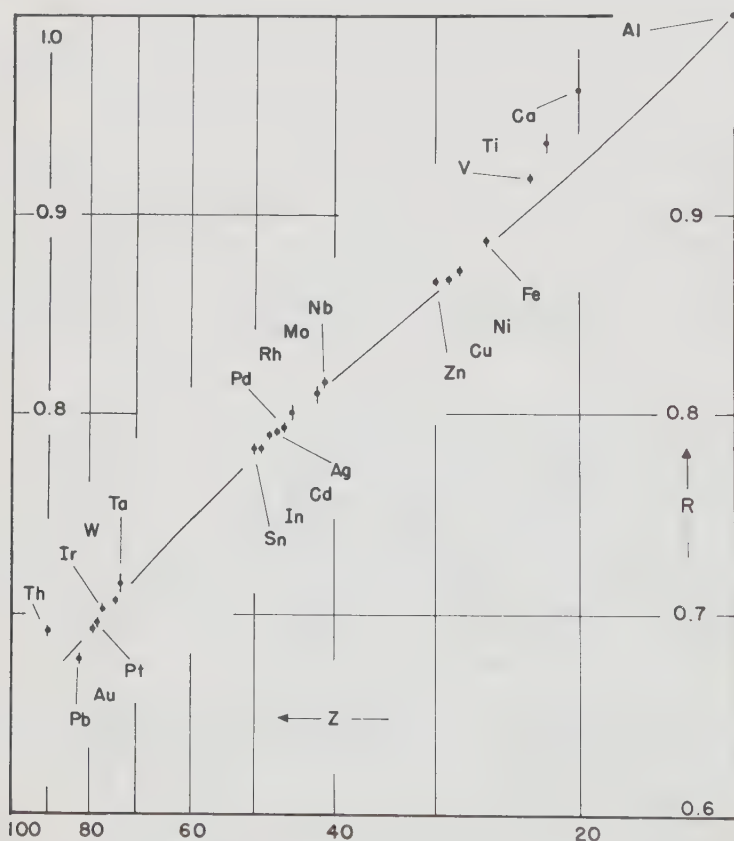


Figure 13. Stopping power as a function of Atomic Number plotted on an expanded scale.

<sup>21</sup>Fig. 13 is Fig. 1 of Ref. 8.

suggestion of a similar effect between Th and Pb. An anomalously high value for the stopping power per electron for Th has also been noted by previous workers. These effects appear to be associated with the systematics of atomic shell structure. Thus, in the range from Ca to Ni the 4s shell is filled and electrons are being added to the inner 3d shell as the succession of elements is built up. Similar shell effects might be expected to show up in the gaps in the data when additional measurements are made and in the neighborhood of Mo (filling of the 4d shell) when more accurate measurements are made.

We can anticipate that there will be further discussion of the valence effects in later sessions of the conference.



### 3. The Dependence of Stopping Power on Chemical Binding

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Recent experiments performed at Nebraska indicate that chemical binding effects may be quite large at low energies. The stopping power of protons and helium ions has been measured in a number of solid hydrocarbons and the results have been compared and analyzed together with similar data from other laboratories on gaseous hydrocarbons. The principal results of this work may be summarized by saying that the effects of the physical state of the hydrocarbon are small and that the effects of chemical binding at energies below 150 kev are large. When proper account is taken of the chemical effects by the introduction of empirical stopping power cross sections for the various bonds appearing in the molecule, all of the data down to proton energies of 40 kev can be correlated within the experimental errors of less than 3 per cent.

The people at Nebraska who have been involved in the experiments are E. J. Zimmerman, D. C. Lorents, and Chester A. Sauter. This work began with an extension of the analyses of measurements made at the California Institute of Technology on gases and with further measurements made by us on solids. The measurements made at CIT<sup>22</sup> were on four hydrocarbon gases ( $\text{CH}_4$ ,  $\text{C}_2\text{H}_4$ ,  $\text{C}_6\text{H}_6$  and  $\text{C}_2\text{H}_2$ ) with protons of 30 to 600 kev energy. Our measurements were on solid  $(\text{CH}_2)_n$ ,  $(\text{C}_8\text{H}_8)_n$  and  $(\text{C}_{12}\text{H}_{14})_n$ , i. e., on polyethylene, polystyrene, and GR-S rubber respectively, using proton energies of 40 to 340 kev. The people at CIT were able to fit their experimental cross sections above 150 kev using a single value of the carbon cross section and a value of the hydrogen atom cross section equal to half the hydrogen molecule cross section. Below 150 kev it was not possible to fit the experimental data in this way. There are, in fact, some outstanding examples of deviations from Bragg's rule. Reynold's data show that the stopping power of 3 ( $\text{C}_2\text{H}_2$ ) molecules is considerably larger than that of

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<sup>22</sup>Reynolds, Dunbar, Wenzel, and Whaling, Phys. Rev. 92, 742 (1953).

one  $C_6H_6$  molecule at energies below 100 kev. This example alone suggests that the chemical binding effects are large.

The measurements on solids were performed in the following manner. The nearly monoenergetic beam of protons or helium ions from the Nebraska Cockroft-Walton ion accelerator was deflected through an angle of  $90^\circ$  in the field of a magnetic spectrometer. A thin film of the stopping material was placed in the path of the beam before the spectrometer. The change in energy of the beam was determined by measuring the decrease in magnetic field necessary to restore the beam to the original  $90^\circ$  deflection angle. The weight per unit area of this film was determined by weighing a known area of the film on a quartz fiber microbalance. The films were sufficiently thin so that the beam sustained only a small fractional energy loss and the measurement was very nearly a true differential stopping power measurement. The experimental results, including also the data on gases from reference 22 as well as Reynold's values for the stopping cross sections of H and C atoms, are given in Table I.

If Reynold's values for  $\sigma(H)$  and  $\sigma(C)$  are used and if it is assumed that  $\sigma(C_m H_n) = m \sigma(C) + n \sigma(H)$ , one can calculate the cross sections for the solid materials and make a comparison with the experimental results. One finds that the agreement is satisfactory above 150 kev but not satisfactory at lower energies. At 100 kev, for example, the cross section for  $CH_2$  is 8 per cent too high, about three times the experimental error. At lower energies, neither the gas data nor the solid data can be fitted in this way.

The hypothesis that the major source of these discrepancies is to be attributed to differences due to physical state of the medium has been examined and discarded. We find instead that all data can be adequately fitted assuming no dependence on the physical state, if we take into account the effects of chemical binding.

One way of taking chemical effects into account is to define different stopping cross sections per electron for the different states of the electron in the hydrocarbon molecule. Each electron of the molecule falls into one of five categories and we define stopping cross sections for each. These are:

- $\sigma_1$  for the H - C bond
- $\sigma_2$  for the C = C bond
- $\sigma_3$  for the C - C bond
- $\sigma_4$  for the C = C bond
- $\sigma_K$  for electrons in the carbon K Shell

TABLE I

Experimental Values of Proton Stopping Cross Sections in Units of  $10^{-15}$  ev  $\text{cm}^2$ /molecule

| Energy<br>kev | CH <sub>4</sub><br>gas | (CH <sub>2</sub> ) <sub>n</sub><br>solid | C <sub>2</sub> H <sub>4</sub><br>gas | C <sub>6</sub> H <sub>6</sub><br>gas | (C <sub>8</sub> H <sub>8</sub> ) <sub>n</sub><br>solid | (C <sub>12</sub> H <sub>14</sub> ) <sub>n</sub><br>solid | C <sub>2</sub> H <sub>2</sub><br>gas | H    | C     |
|---------------|------------------------|------------------------------------------|--------------------------------------|--------------------------------------|--------------------------------------------------------|----------------------------------------------------------|--------------------------------------|------|-------|
| 30            | 37.4                   | 22.5                                     |                                      | 116.0                                | 149.3                                                  |                                                          | 43.4                                 | 5.84 |       |
| 40            | 39.7                   | 24.2                                     | 54.4                                 | 126.0                                | 160.9                                                  |                                                          | 47.4                                 | 6.25 |       |
| 50            | 40.9                   | 25.2                                     | 57.4                                 | 133.0                                | 168.2                                                  | 253.4                                                    | 49.5                                 | 6.43 |       |
| 60            | 41.3                   | 25.8                                     | 58.7                                 | 135.7                                | 171.8                                                  | 264.2                                                    | 49.8                                 | 6.45 |       |
| 70            | 41.2                   | 26.1                                     | 58.8                                 | 135.5                                | 173.2                                                  | 268.9                                                    | 49.2                                 | 6.36 |       |
| 80            | 40.8                   | 26.1                                     | 58.0                                 | 134.4                                | 172.5                                                  | 270.0                                                    | 48.0                                 | 6.23 |       |
| 90            | 40.0                   | 25.7                                     | 56.7                                 | 133.5                                | 171.5                                                  | 268.7                                                    | 46.7                                 | 6.04 |       |
| 100           | 38.9                   | 25.2                                     | 55.5                                 | 131.7                                | 169.0                                                  | 265.2                                                    | 45.0                                 | 5.83 | 16.25 |
| 125           | 36.2                   | 23.8                                     | 52.3                                 | 124.3                                | 158.3                                                  | 250.5                                                    | 41.8                                 |      |       |
| 150           | 33.6                   | 25.2                                     | 48.8                                 | 116.5                                | 148                                                    | 234.2                                                    | 38.5                                 | 4.70 | 14.6  |
| 175           | 31.0                   | 21.2                                     | 45.0                                 | 109.0                                | 137.2                                                  | 219.0                                                    | 35.6                                 |      |       |
| 200           | 28.6                   | 19.9                                     | 41.4                                 | 102.0                                | 127.6                                                  | 206.4                                                    | 32.9                                 | 3.90 | 12.7  |
| 225           | 26.6                   | 18.7                                     | 38.6                                 | 95.2                                 | 119.1                                                  | 195.1                                                    | 30.9                                 |      |       |
| 250           | 24.8                   | 17.6                                     | 36.1                                 | 89.2                                 | 111.3                                                  | 184.9                                                    | 29.0                                 | 3.33 | 11.28 |
| 275           | 23.2                   | 16.6                                     | 34.0                                 | 84.3                                 | 104.4                                                  | 175.7                                                    | 27.4                                 |      |       |
| 300           | 22.0                   | 15.7                                     | 32.0                                 | 79.8                                 | 99.9                                                   | 166.8                                                    | 25.9                                 | 2.91 | 10.20 |
| 325           | 20.8                   | 15.1                                     | 30.4                                 | 75.8                                 | 95.2                                                   | 158.6                                                    | 24.6                                 |      |       |
| 350           | 19.8                   | 14.6                                     | 28.8                                 | 72.2                                 | 90.4                                                   | 151.0                                                    | 23.5                                 | 2.60 | 9.30  |
| 400           | 18.1                   |                                          | 26.2                                 | 66.3                                 |                                                        |                                                          | 21.4                                 | 2.35 | 8.54  |
| 450           | 16.65                  |                                          | 24.1                                 | 61.4                                 |                                                        |                                                          | 19.7                                 | 2.14 | 7.94  |

If we use coefficients  $a$ ,  $b$ , and  $c$  to denote the number of double, single, and triple carbon to carbon bonds in the molecule and neglect the contribution to stopping of the nuclei themselves at these low energies, the expression for the molecule stopping cross section is  $\sigma(C_m H_n) = 2n\sigma_1 + 4a\sigma_2 + 2b\sigma_3 + 6c\sigma_4 + 2m\sigma_K$ .

The five cross sections appearing in this equation are to be determined from the experimental data on the molecular cross sections. If the hypothesis is valid, one set of values should be adequate to describe all of the data.

In applying this equation to the data of Table I we make little error if we neglect  $\sigma_K$  entirely. The numerical value of  $\sigma_K$  may be determined from the curve for the stopping power of K electrons as given in the first paper of reference 19. From the stopping number  $B_K$  one calculates the stopping cross section and finds that for carbon this is  $0.24 \times 10^{-15}$  ev cm<sup>2</sup>/electron at 200 kev and  $0.30 \times 10^{-15}$  ev cm<sup>2</sup>/electron at 300 kev. These numbers are sufficiently small in comparison with the measured values of  $\sigma(C_m H_n)$  below 200 kev to be unimportant in a first analysis. Consequently, there are four cross sections to be determined from the data on seven compounds. For six of these compounds the coefficient  $c = 0$ . This leaves three parameters to be determined by six sets of experimental data. The fourth parameter is then determined by the data on the seventh compound.

The least squares values of  $\sigma_1$ ,  $\sigma_2$ ,  $\sigma_3$  and  $\sigma_4$  determined in this way are shown in Figure 14. When the cross sections of Figure 14 are inserted into the equation for  $\sigma(C_m H_n)$  one obtains cross sections which agree within the experimental errors for all compounds at energies below 200 kev. Above 200 kev the agreement is good except for deviations of as much as 3 per cent in the case of  $C_8H_8$  and  $C_{12}H_{14}$ , one experimental curve lying higher and the other lower than the calculated curve. If  $\sigma_K$  had been taken into account the values of  $\sigma_1$  would have been lower by 0.08 and the values of  $\sigma_2$ ,  $\sigma_3$  and  $\sigma_4$  would have been lower by 0.15 at 300 kev. At lower energies the reduction would have been smaller.

For proton energies between 40 and 200 kev the molecular cross sections calculated in this way are probably more accurate than those calculated by other methods and on the basis of other models. However, it would be desirable to repeat the measurements on  $C_8H_8$  and  $C_{12}H_{14}$  above 200 kev to see whether the discrepancies which we have noted are real. Also, the present method



of analysis should be checked further by using data on other hydrocarbon compounds when it becomes available.

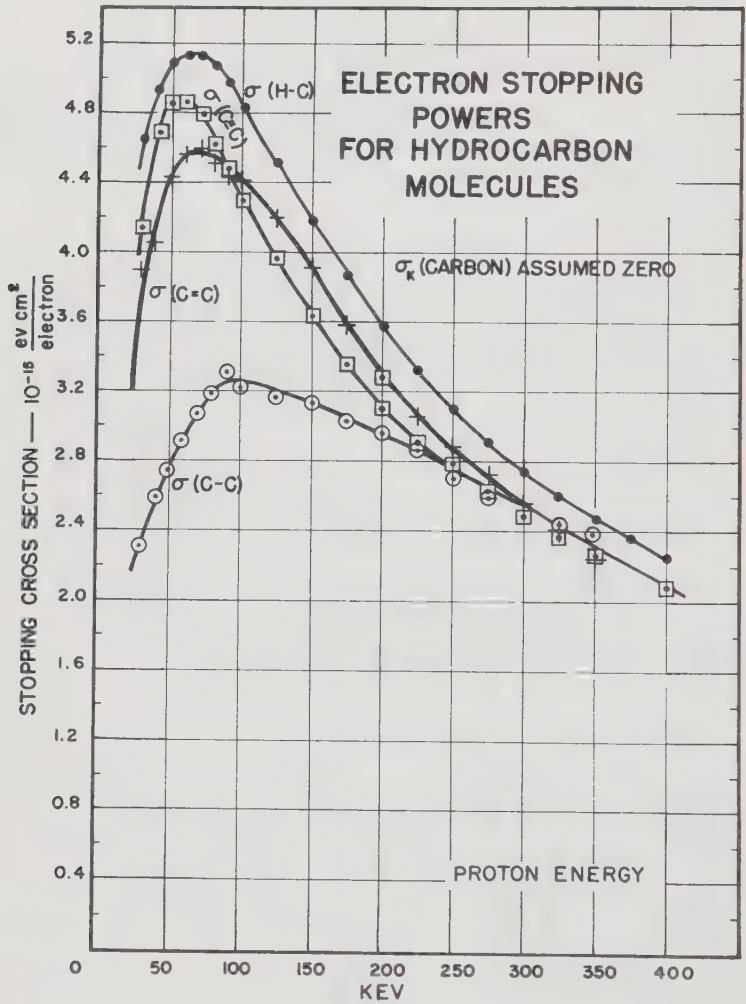


Figure 14. Stopping power of electrons in different bonds of the hydrocarbon molecule.

#### 4. Range Measurements

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University of Washington

The ranges of charged particles in matter have been studied extensively. The data themselves are a primary objective in most of these studies because extensive range-energy information covering a number of different particles, a wide variety of stopping materials, and a wide range of energies are of the greatest importance as an aid in experimental work; e. g., in the study of nuclear reactions, the identification of particles, etc. An index to all of the information on ranges, as well as on stopping powers, which has become available up to early 1957 shows how extensive this information now is.<sup>23</sup>

Our problem, however, is a different one. We are concerned here with the use of experimental information for the purpose of theoretical evaluation, to be followed possibly by theoretical extrapolation into areas where experimental data is unavailable. We find that from this point of view most of the data is of little use to us. Some of it is simply of insufficient accuracy. Also we are forced to accept the restriction that the theory itself is not wholly adequate. As a consequence of limitations in the theory we can expect to obtain reliable conclusions at this stage in the development of our subject only if we restrict our attention to certain particle types and to ranges of particle energies in which the theory has the least ambiguity. Thus we eliminate at this stage all of the range data on alpha particles of energies less than 8 Mev. Similarly we consider protons whose energies are not lower than about 1.25 Mev. These restrictions reduce the amount of data considerably. The

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<sup>23</sup>Index and Annotated Bibliography of Range and Stopping Cross Section Data, LA-2156, compiled by Ronald E. Brown and Nelson Jarmie, Los Alamos Scientific Laboratory (1957), available from the Office of Technical Services, U. S. Department of Commerce, Washington 25, D. C.

following items dealing solely with protons and including some very recent measurements appear to be most useful:

Parkinson, Herb, Bellamy and Hudson, Phys. Rev. 52, 75 (1937). Absolute extrapolated ranges for protons in aluminum and air, 0.1 to 1.8 Mev. Accuracy 10 to 3 per cent.

R. R. Wilson, Phys. Rev. 60, 749 (1941). Mean ranges up to 4 Mev, relative to air. Accuracy 2 per cent.

Bloembergen and Van Heerden, Phys. Rev. 83, 561 (1951). Absolute mean ranges 35 - 75 Mev. Accuracy about  $\pm 1$  per cent.

Mather and Segré, Phys. Rev. 84, 191 (1951). Absolute mean ranges at 240 Mev. Accuracy about  $1/2$  per cent.

D. H. Simmons, Proc. Phys. Soc. (London) A65, 454 (1952). Mean range in Al at about 10 Mev relative to range in emulsion. Accuracy  $\sim 1$  per cent.

Hubbard and MacKenzie, Phys. Rev. 85, 107 (1952). Absolute mean range in Al at 18.0 Mev. Accuracy 0.2 per cent.

Bichsel, Mozley, and Aron, Phys. Rev. 105, 1788 (1957). Absolute mean ranges 6 to 18 Mev. Accuracy  $\sim 0.3$  per cent.

H. Bichsel, Phys. Rev. 112, 1089 (1958). Absolute mean range in 1 to 6 Mev. Accuracy 1 to 0.3 per cent.

Barkas et al., Nuovo Cim. VIII, 185 (1958). Measurements from 1 to 1000 Mev equivalent proton energy in emulsions described and discussed. High accuracy  $\sim 1/2$  per cent.

Our interest in range measurements stems from the relatively high accuracy with which these measurements can be made. At first sight it will appear that from the point of view of the evaluation of parameters, in the stopping power relations range measurements might be less satisfactory than stopping power measurements since they are one step further removed from a direct

comparison with theory. However, the accuracy of measurement is such that range measurements as a technique for numerical evaluation does compare favorably with the use of stopping power measurements for the same purpose.

In order to obtain an accurate range-energy measurement one must have:

- (a) Accurate determinations of energy.
- (b) Accurate measurements of foil thickness.
- (c) An accurate plot of the transmission curve in the neighborhood of the mean range and for some distance into the region of nearly full transmission.

These matters are discussed in a report on range-energy measurements for protons of 6 - 18 Mev energy in a number of metals.<sup>24</sup> The experimental procedures used in these measurements differ in several respects from those used before. Instead of having protons of a fixed energy impinge on a stacked foil, protons of variable energy impinge on a single foil. The transmission curve is plotted and the energy  $E_0$  corresponding to a mean range equal to the absorber thickness is obtained as the energy at which half the protons are transmitted. The energies are measured by the deflection of the proton beam in the field of a stabilized electromagnet using nuclear induction methods for the measurement of field and several different geometries for definition of the beam to provide an internal check on the measurements. A triple coincidence proportional counter was used to give reliable detection of protons. The over-all errors in these measurements are estimated to be 0.14 per cent in the energy determination and 0.1 per cent in the range determination.

In order to compare the results of range energy measurements with the theory of stopping power we must integrate the stopping power relation to obtain a predicted range, or we differentiate the experimental range-energy relation. As we will show the latter procedure is a good one if sufficient data are available to define an

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<sup>24</sup>H. Bichsel, R. F. Mozley, and W. A. Aron, Phys. Rev. 105, 1788 (1957).



analytical curve. For the present we will proceed according to the first method. The mean range at the energy  $E$  is

$$R(E) = R(E_0) + \int_{E_0}^E \frac{dE}{(-dE/dR)}$$

when  $E_0$  is chosen to be the lowest energy at which a reliable range determination has been made and which is at the same time not so low that the theoretical stopping power formulae cannot be used. Tables of  $R$  as a function of  $E$  are constructed and compared with the experimental results.

One of the difficulties in making this comparison is that the theoretical expression for  $(-dE/dR)$  depends on the mean ionization potential  $I$  which is to be determined by the data and on binding energy corrections which are only partially known. In general we must regard  $I$  and the binding energy corrections for the various electron shells ( $C_K, C_L, \dots$ ) as unknown parameters which must be adjusted until a set of range energy relations consistent with all of the experimental data are obtained. Thus, the comparison with experiment is made by constructing many range energy tables and finally choosing the one which most nearly fits the experimental data. To the extent that this relation may be regarded as a valid relation, one can say the parameters which enter into it have been correctly evaluated.

The stopping power relation which we use in this computation is:

$$\frac{1}{\rho} \left( -\frac{dE}{dR} \right) = \frac{K(\beta)}{A} \left[ Z \left\{ f(\beta) - \ln I \right\} - \sum_i C_i \right]$$

where

$$K(\beta) = \frac{4\pi e^4 N_0}{mc^2 \beta^2} \quad f(\beta) = \ln(2mc^2 \beta^2) - \ln(1 - \beta^2) - \beta^2$$

and  $A$ ,  $Z$ , and  $\rho$  are the atomic weight, atomic number, and the density of the stopping material respectively, and  $N_0$  is Avogadro's number. We then make tables of  $E$ ,  $\beta$ ,  $K(\beta)$ , and  $f(\beta)$  and then with suitable choices of  $I$  and the binding energy corrections  $C_i$  we tabulate the stopping power and perform the integration to obtain the range.

There is one further step which must be taken before the comparison with experimental data is made. The numerical calculation which we have just discussed leads to a determination of the mean range. This is, however, not precisely what we have measured. Multiple scattering and perhaps also nuclear scattering plays

a role in the stopping process, and the thickness of our foil must be regarded as a projected range. Therefore, we must make a multiple scattering correction, and at high energies we should probably have to make a nuclear scattering correction. The calculation of this correction is discussed in reference 24 and elsewhere and we will not discuss it further here. As shown in reference 24 this correction may amount to as much as 5 per cent in a heavy element like gold at energies as low as 10 Mev.

The measurements made at Princeton with 6 to 18 Mev protons have been subsequently extended with comparable precision to 1 Mev using the electrostatic generator at the Rice Institute.<sup>25</sup> With the addition of the new data it is possible to give a reliable analytical relation for  $R(E)$  which expresses the results of the measurements over an extended energy range. For protons in Al the relation given by

$$\begin{aligned} R &= 3.837 E^{1.5874} & 1 < E < 2.7 \text{ Mev} \\ R &= 2.837 E^2 / (0.68 + \log_{10} E) & 2.7 < E < 20 \text{ Mev} \end{aligned}$$

agrees with the experimental results within 0.2 per cent above 3 Mev and within 0.4 per cent below 3 Mev. It is interesting to compare these results with other measurements that we have mentioned previously. The comparison with the measurements of Parkinson, Simons, and Hubbard and MacKenzie, all of whose measurements lie in the range of energy we are considering, and with the measurements of Bloembergen and Van Heerden which lie in a somewhat higher range of energies is shown in Figure 15. The comparison is made by referring all data to our analytical curve.

We have used the data represented by the analytical curve to determine the best possible value of  $I$  for Al. As we have already mentioned this cannot be done without simultaneously determining the binding energy corrections. Since we are in an energy region where both K shell and L shell corrections have some importance, the failure to include these corrections would in principle make the determination of a velocity independent  $I$  impossible. This is in fact the case, as our calculations show. We proceed as follows. The K shell corrections are well known for Al<sup>19</sup>. The theory of L shell corrections is also known for the heavier elements but numerical application of this theory to an element as light as Al is difficult.

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<sup>25</sup>Hans Bichsel, Phys. Rev. 112, 1089 (1958).

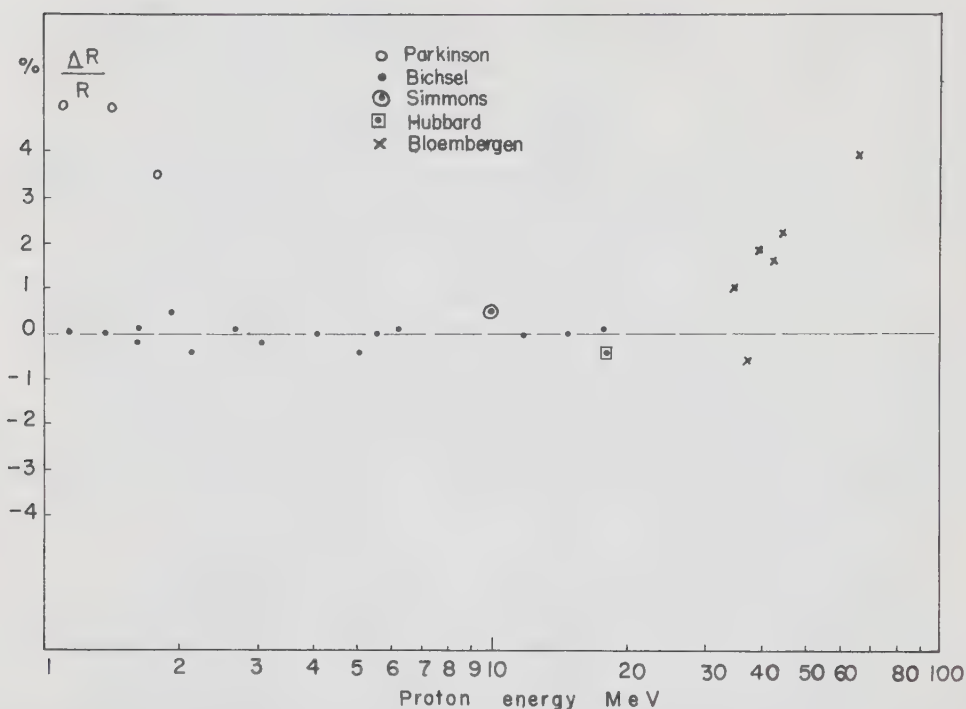


Figure 15. Comparison of Range data for protons obtained by different experimenters.

Our procedure therefore is to take the K shell correction  $C_K(E)$  from Walske's theory, but to permit our experimental data to determine the L shell correction in such a way that  $I$  is constant. We use the theory of the L shell correction only to the extent that we assert that to sufficient accuracy for our purpose  $C_L$  is given by  $C_L = k/E$  for  $E > 4$  Mev. Thus, we are permitting the experimental data to determine the constant  $k$  to the extent that this data is sufficiently accurate to make such a determination. For  $E < 4$  Mev we obtain a tabulation of  $C_L$ .

The results lie between  $I = 164$  ev with  $k = 1.1$  and  $I = 162.8$  ev with  $k = 1.6$ . These results for  $k$  agree with the expectation based on the theory of Walske and the value of  $I$  is consistent with the value obtained from the absolute stopping power measurement of Sachs as has been mentioned in a previous report by MacKenzie. However, our calculation still requires some further refinement, particularly with regard to the multiple scattering correction. The final result is not expected to differ much from the results just given, however.

Our results for elements heavier than Al, e. g., Ni and Ag, are in a more preliminary stage. For these elements both the K shell and the L shell corrections can be taken from the theory, and it is now the M shell correction which we try to determine from experiment. Following the same procedure as before we express the M shell correction in the asymptotic form  $C_M = k'/E$  and let experiment determine the constant  $k'$ . It may be of some interest in this case to tabulate our results. These are as follows:

| $E^P$<br>(Mev) | $dE/dR$<br>(Mev cm <sup>2</sup> /gm) | $C_K + C_L$ | $Z \ln I$<br>(for $C_M=0$ ) | $Z \ln I$<br>(for $C_M=22/E$ ) |
|----------------|--------------------------------------|-------------|-----------------------------|--------------------------------|
| 2.0            | 82.5                                 | 2.7         | 164.5                       | 153                            |
| 2.5            | 72.7                                 | 3.4         | 163                         | 154                            |
| 3.0            | 65.3                                 | 3.8         | 163                         | 156                            |
| 3.5            | 59.2                                 | 3.9         | 162                         | 156                            |
| 4.0            | 54.1                                 | 4.0         | 162                         | 156                            |
| 4.5            | 49.7                                 | 4.0         | 163                         | 157                            |
| 5.0            | 45.9                                 | 4.0         | 163                         | 159                            |
| 12.0           | 25.2                                 | 3.0         | 160                         |                                |
| 19.0           | 17.72                                | 2.4         | 162                         | 161                            |

All of the low energy stopping power data in this table are obtained by differentiation of our range energy curves. The one point at 19 Mev is taken from reference 8 after normalizing to our absolute Al values.

The last two columns of the table representing values of  $Z \ln I$  show that we obtain the best results if the magnitude of the  $C_M$  correction is minimized. In fact on the basis of this information there is little reason to choose an M shell correction greater than  $C_M = 4/E$  in this energy range. This is somewhat smaller than one would expect on the basis of Walske's theory, but our results in the absence of a better application of the multiple scattering correction must still be regarded as preliminary. Choosing  $Z \ln I = 163$  for  $C_M = 0$  we obtain  $I = 337$  ev, corresponding to  $I/Z = 12.0$ .



The major unsolved problems are the consistent application of the multiple scattering correction and the better evaluation of  $C_M$  and higher shell corrections.<sup>26</sup>

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<sup>26</sup>Some results on the application of the multiple scattering correction with particular reference to new data on Ag as well as on Ni have been obtained since the conference. These results will be published separately. However, the results on Ag are of particular interest and will be noted here. Using Walske's  $C_M$  and  $C_L$  corrections we obtain a velocity independent  $I$  equal to 510 ev if we take  $C_M$  asymptotically equal to  $(20 \pm 10)/E$ . This value of  $I$  is considerably lower than the value of 585 ev which we quoted in ref. 24, and it is somewhat higher than the value of 462 ev obtained from the Bakker-Segré data when the  $A_1$  value is normalized to 164 ev.

## 5. Range Measurements of Singly and Multiply Charged Particles in Emulsion

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University of California at Berkeley

Nuclear track emulsion is at present the substance in which it is most important that energy loss rates of charged particles be known. All the meson and strange particle masses and reaction energies have been determined in emulsion or are tied directly to emulsion measurements. I regard the range-energy curve for standard emulsion as the most important calibration relation in high energy physics. We need a reliable curve extending from proton energies above a billion electron volts down to a few hundred kilovolts because the relation is required on the one hand to determine energies of extremely high-velocity strange-particle secondaries, and, on the other, to determine energies of slow multiply-charged disintegration fragments.

Because range measurements are of such paramount importance for quantitative emulsion work, we have given much attention to the establishment of range-energy relations for emulsion. To do this we have had to consider nearly all the classical problems of energy loss of charged particles, as well as several that are peculiar to emulsion.

The general experimental problems are:

- (a) How to obtain particle beams of many velocities free of interfering background.
- (b) How very accurately to know the particle velocities.
- (c) How to avoid errors arising from scattering.
- (d) How to carry out the range measurement with minimum error.

Here emulsion has a decided advantage over most other absorbers because the whole trajectory of each particle as it penetrates the

stopping medium is witnessed and in principle can be measured with almost arbitrary accuracy. The angles of entrance of each particle into the emulsion also can be measured, and these may be necessary for precise momentum determination:

The general theoretical problems are:

- (a) The question of the existence of and the evaluation of a constant mean ionization potential.
- (b) The evaluation of corrections to the Bethe-Bloch theory at high and low velocities.
- (c) The complications in the theory of stopping of slow multiply-charged ions.
- (d) The effects of nuclear interactions in thick absorbers.

The problems peculiar to emulsion are:

- (a) The establishment of a standard density and composition for emulsion.
- (b) To provide correction procedures for conversion of ranges to standard conditions when measured in non-standard emulsion.
- (c) To evaluate distortion effects and range straggling effects not encountered in a rigid homogeneous medium.
- (d) To evaluate errors and corrections arising from the necessity of tracing tracks from one pellicle to another in an emulsion stack.

One of the most important experimental techniques we have employed is  $180^\circ$  bending of particles in the internal magnetic field of the cyclotron. The type of apparatus used has been described by

us before.<sup>27, 28, 29</sup> As the particles move in the radially decreasing field of the cyclotron the orbits precess and a rather elaborate treatment is required in order to make a precise determination of the momentum.<sup>28</sup> Using the procedure we have developed, the momenta can be determined to one part in 1000 in absolute value. The general type of result obtained when range is plotted against magnetic rigidity is shown in Figure 16. The results of extensive careful measurements on fast singly-charged particles have been reported in a recent article.<sup>27</sup>

I have calculated also a completely theoretical range-energy relation in which the mean ionization potential is the only adjustable parameter.<sup>30</sup> The principal quantities to be discussed are contained in the stopping power relation:

$$\left( - \frac{dE}{dR} \right) = \frac{4\pi z^2 e^4 N Z}{m c^2 \beta^2} \left[ \ln \frac{2 m c^2 \beta^2}{I} - \ln(1 - \beta^2) - \beta^2 - C(\beta) \right]$$

where  $ze$  is the charge of the incident particle,  $NZ$  is the number of electrons per unit volume of the medium,  $m$  is the electron mass,  $\beta = v/c$ ,  $I$  is the mean ionization potential, and  $C(\beta)$  is a correction which takes into account the binding energy corrections at low energies and the polarization corrections at high energies. At low energies where the polarization correction is velocity independent and  $I$  is defined to be the constant effective mean ionization potential,  $C(\beta)$  contains only the binding energy corrections and may be written as

$$C(\beta) = \frac{1}{Z} \sum_i C_i = \frac{1}{Z} (C_K + C_L + \dots)$$

<sup>27</sup>Barkas, Barrett, Cuer, Heckman, Smith, and Ticho, the Range-Energy Relation in Emulsion, Part 1, UCRL-3768, University of California (1957) and *Nuovo Cimento* 8, 185 (1958).

<sup>28</sup>Barkas, Birnbaum, and Smith, *Phys. Rev.* 101, 778 (1956). See Fig. 1 of this ref. and the accompanying discussion in regard to the momentum determination.

<sup>29</sup>L. E. Bailey, University of California Radiation Laboratory Report UCRL 3334 (1956).

<sup>30</sup>Walter H. Barkas, The Range-Energy Relation in Emulsion, Part 2, UCRL-3769, University of California (1957), and *Nuovo Cimento* 8, 201 (1958).



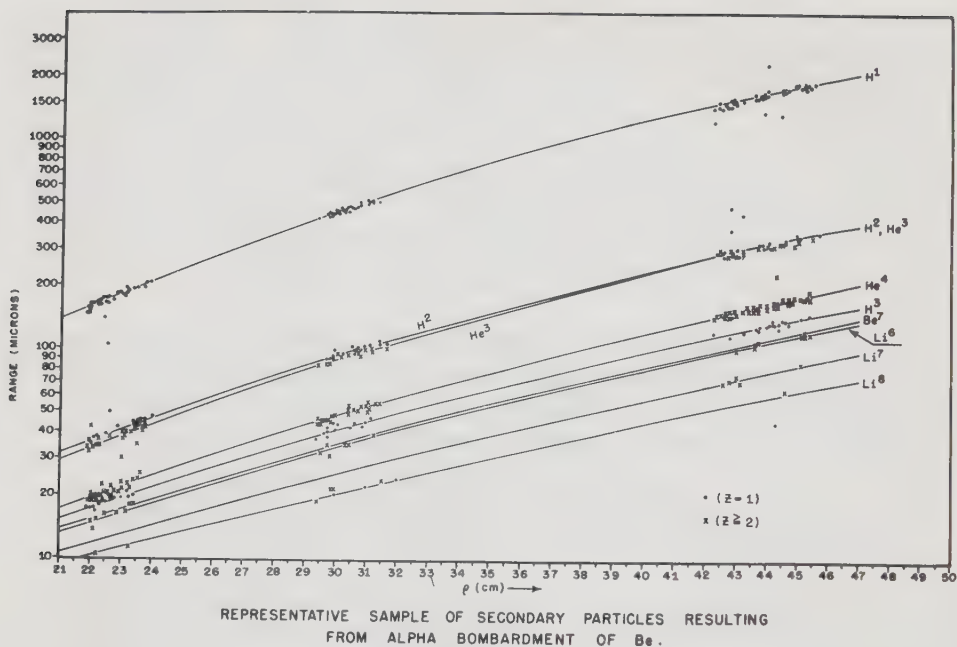


Figure 16. Isotope Loci on Range-Radius of Curvature diagram. Each point represents a track found on one of three plates placed at various distances from a small internal target in the 184-inch cyclotron. The radius of curvature,  $\rho$ , of the particle trajectory in the magnetic field was calculated from the position and direction of the track in the emulsion. The range also was measured. The grain density of the track revealed whether the particle was singly or multiply charged. The solid lines are calculated loci of points corresponding to the isotopes listed.

where  $C_i$  is the binding energy correction of the  $i^{\text{th}}$  shell as defined by Walske.<sup>19</sup> At high energies we must include the effects of polarization in the determination of  $C(\beta)$ . The latter have been most thoroughly investigated by Sternheimer.<sup>31</sup> Numerical values of  $C(\beta)$  for standard emulsion as calculated by us on the basis of the theories of Walske and Sternheimer are tabulated in Table II. The binding energy corrections appearing in this tabulation of  $C(\beta)$  include the L shell corrections for iodine, silver, and bromine as well as the K shell corrections for all elements (except hydrogen), but the more uncertain M shell corrections are not included.

<sup>31</sup>R. M. Sternheimer, Phys. Rev. 103, 511 (1956).

TABLE II

| E in Mev | C      | E in Mev | C     | E in Mev | C     |
|----------|--------|----------|-------|----------|-------|
| 1.2      | -0.081 | 5.0      | 0.072 | 200      | 0.016 |
| 1.4      | -0.059 | 5.4      | 0.075 | 260      | 0.011 |
| 1.6      | -0.040 | 6.0      | 0.079 | 300      | 0.009 |
| 1.8      | -0.023 | 7.0      | 0.084 | 400      | 0.006 |
| 2.0      | -0.009 | 8.0      | 0.085 | 500      | 0.005 |
| 2.2      | +0.003 | 9.0      | 0.086 | 700      | 0.004 |
| 2.4      | 0.014  | 10.0     | 0.088 | 1,000    | 0.002 |
| 2.6      | 0.023  | 12.0     | 0.087 | 1,200    | 0.004 |
| 2.8      | 0.031  | 16.0     | 0.083 | 1,400    | 0.009 |
| 3.0      | 0.038  | 20.0     | 0.078 | 1,600    | 0.020 |
| 3.2      | 0.043  | 24.0     | 0.073 | 1,800    | 0.033 |
| 3.4      | 0.048  | 28.0     | 0.067 | 2,000    | 0.046 |
| 3.6      | 0.053  | 30.0     | 0.065 | 3,000    | 0.115 |
| 3.8      | 0.057  | 40.0     | 0.055 | 4,000    | 0.184 |
| 4.0      | 0.060  | 50.0     | 0.048 | 5,000    | 0.250 |
| 4.2      | 0.063  | 70.0     | 0.038 | 10,000   | 0.524 |
| 4.4      | 0.065  | 100.0    | 0.030 | 20,000   | 0.902 |
| 4.6      | 0.067  | 140.0    | 0.023 | 30,000   | 1.167 |
| 4.8      | 0.070  |          |       |          |       |

The purely theoretical curve obtained on this basis and the comparison with experimental results is shown in Figure 17. It is seen that, although the fit is generally good, some irregularities appear at low energies that are outside the limits of experimental error. It is found that these irregularities are greatly decreased for protons if below 40 Mev one merely permits a smooth deviation from the Bethe-Bloch theory. The existing K and L shell corrections (as derived by Walske) were based on hydrogen-like wave functions and, even in the absence of M shell corrections, they appear to overemphasize the shell effects at low velocities. If a smooth deviation from the Bethe-Bloch theory is permitted between 5 and 40 Mev, and below 5 Mev an empirical curve is used, we obtain a range-energy relation (Figure 18) that fits our data rather well.

The evaluation of the mean ionization potential applicable to emulsion has been made by trial integration of the Bethe-Bloch formula using various values of the mean ionization potential.

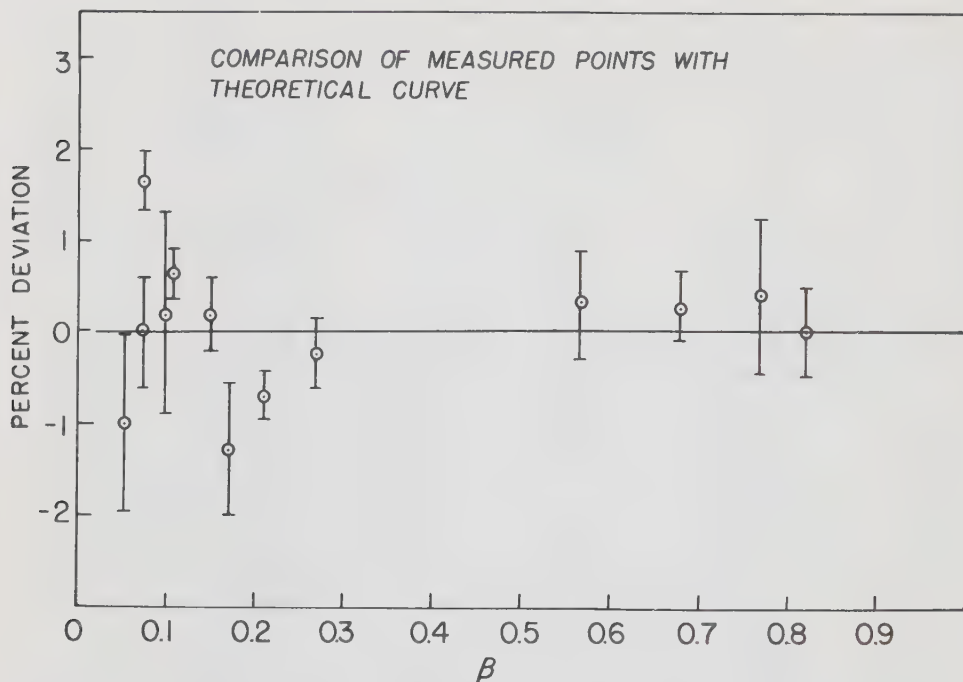


Figure 17. Deviation of the measured range points from the purely theoretical curve. At low velocities some points deviate more than is likely with the known errors, and the form of the deviation is of a sort that would be produced by over correction for shell effects. A smoother correction curve appears to be needed.

In obtaining the best value most weight was given to the points at high velocity where the correction term  $C$  was almost negligible. The final value found was  $I = 331 \pm 6$  ev.

This corresponds to a mean value of  $I/Z$  for emulsion atoms exclusive of hydrogen equal to  $12.1 \pm 0.2$  ev. Since the electrons of the light atoms constitute only 18 per cent of the total, this figure should be close to the ratio  $I/Z$  appropriate to silver and bromine, the most important constituents of emulsion. Of course, we cannot readily study  $I/Z$  as a function of  $Z$  in emulsion, but it does appear that at high velocities the Bethe-Bloch theory with a fixed value of the mean ionization potential is capable of describing the dependence of the rate of energy loss on the velocity as accurately as it can now be measured.

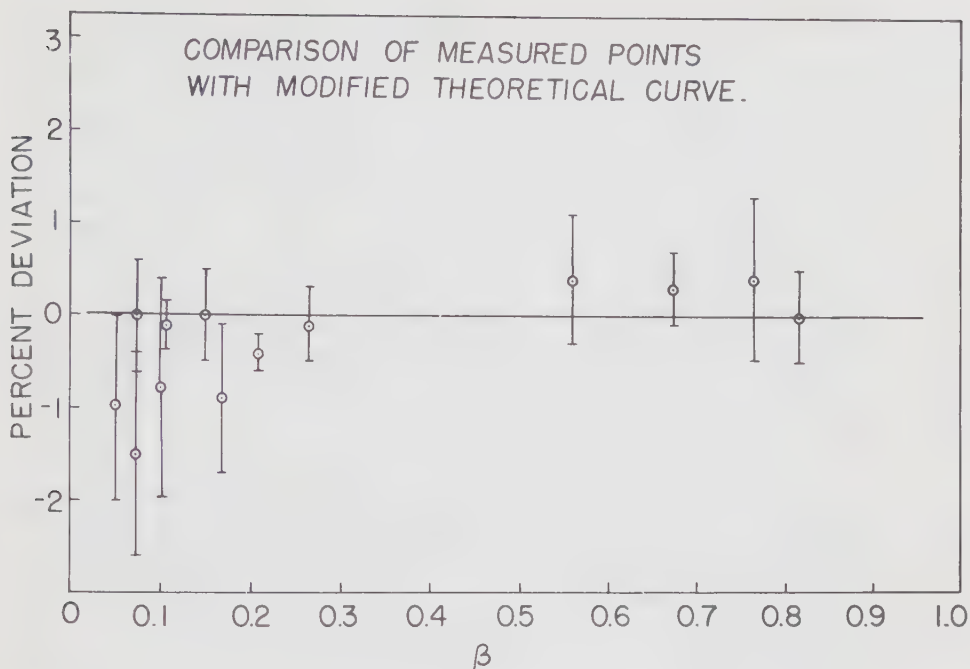


Figure 18. Comparison of our measured points with the adopted range-energy relation. Theory was used to establish the mean ionization potential from our empirical ranges for  $\beta > 0.27$ . At lower velocities a smooth deviation from the Bethe-Bloch theory was permitted, and other data than our own were considered.

While we report our ranges as those of protons, the data is applicable to many kinds of particles, and in the determination of the range curve we have measured ranges of  $\mu$ ,  $\pi$ , and K mesons, protons, deuterons, and tritons, as well as of helium-3 and helium-4 nuclei. The connection between the range of an ideal proton and the actual measured range,  $R_1$ , between the extremities of the first and last track grains is the following:

$$R_1 = \frac{M}{z^2} (\lambda + B_z) f(\rho, \beta) + \epsilon = R f(\rho, \beta) + \epsilon$$

Here  $R$  is the corrected range in standard emulsion, the quantities  $M$  and  $z$  are the mass and nuclear charge of the particle or ion in units of the proton,  $\lambda$  is the ideal proton range for velocity  $\beta c$ ,  $B_z$  is a correction which allows for the extension of range of a positive ion caused by the neutralization of its charge by electrons that it



captures at low velocities,  $f(\rho, \beta)$  is a function of the emulsion density,  $\rho$ , and, to some extent, of the velocity. It falls as  $\rho$  increases and is unity for standard emulsion of specific gravity 3.815, whatever the velocity. As the velocity increases, the function  $f$  becomes more nearly inversely proportional to  $\rho$ . The quantity  $\epsilon$  corrects for effects caused by the granularity of the emulsion and for end effects that depend on the emulsion sensitivity. These are small and too detailed to discuss here. By the ideal proton-range,  $\lambda$ , is meant the mean range in a homogeneous material having the composition of standard emulsion of a particle having the proton mass and charge but which does not capture electrons at the end of its range like a positive proton, nor, like a negative proton, have a high probability itself of being captured before coming to rest.

In order to evaluate the density  $\rho$ , I treat non-standard emulsion as a linear combination of standard emulsion, silver halide, water and loading material. Then  $\rho$  is expressed in terms of the concentrations (zero, positive, or negative) of these four components. For practical purposes this seems adequate to care for our immediate needs.

In our published study we made no measurements on very slow particles, and low-velocity emulsion range-data from the literature are difficult to interpret because often the emulsion density is poorly known, the mean grain spacing and mean gap length are not given, and the precise method of making the end corrections is seldom discussed. Recently, in order to evaluate the range at low velocities, we have made use of differential energy loss measurements that have been made in pure elements.

Dr. Ward Whaling has kindly supplied a preprint of his recent report on "The Energy Loss of Charged Particles in Matter."<sup>32</sup> The atomic stopping powers are found to be relatively smooth functions of the atomic number except for very low velocities.

From these data a curve of rate of energy loss versus velocity of protons has been constructed for material with the composition of emulsion. The presence of many elements in emulsion smooths and averages the curve. The range curve is found from these data

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<sup>32</sup>W. Whaling, Encyclopedia of Physics, Springer - Verlag, Berlin, 34, 208 (1958).

by integration. The constant of integration,  $\epsilon$ , must be determined from measurements on particle tracks, and an analysis of the process of grain formation. This has been done by measuring the range of protons from the reaction  $N^{14}(n, p) C^{14}$  in G. 5 and C. 2 emulsion. The details are too lengthy to mention here, but the results are given in Table III.

TABLE III

Energy Loss Rate and Range in Standard Emulsion

| $\gamma$ (Mev)            | 0.1  | 0.2  | 0.3  | 0.4  | 0.6  | 1.0   | 1.5   | 2.0   | 2.5   |
|---------------------------|------|------|------|------|------|-------|-------|-------|-------|
| $\delta$ (Mev/cm)         | 1500 | 1130 | 950  | 820  | 650  | 490   | 380   | 320   | 270   |
| $\lambda$ ( $10^{-4}$ cm) | 0.99 | 1.78 | 2.76 | 3.91 | 6.69 | 13.92 | 25.63 | 39.98 | 57.06 |

The quantity  $\gamma$  is the kinetic energy in Mev, and  $\lambda$  is the range of an ideal proton. Then  $\delta (\equiv d\gamma/d\lambda)$  is the energy loss per centimeter of an ideal proton. For a nucleus of charge  $z$  and mass  $M$  in units of the proton, the kinetic energy,  $T$ , is  $M\gamma$  and the corrected range,  $R$ , is equal to  $M(\lambda + B_z)/Z^2$ .

The range curve joins smoothly to that previously established at higher velocities.

We have also studied the range straggling, including not only the directly calculable Bohr straggling in a rigid homogeneous medium, but also the several straggling effects that arise because of distortions and heterogeneity of the stopping medium. A typical straggling curve is shown in Figure 19.

Recently we have turned to the problem of multiply-charged ions that are not fully stripped of electrons. We did some work along these lines several years ago by comparing the ranges, in the same emulsion plate, of readily identifiable  $B^8$  and  $Li^8$  nuclei with hydrogen and helium ranges. The data were used to study the

quantity  $B_z = \frac{Z^2 R}{M} - \lambda(v)$  which measures the extension of range produced by the tendency of multiply charged nuclei to be neutralized by electrons they capture.

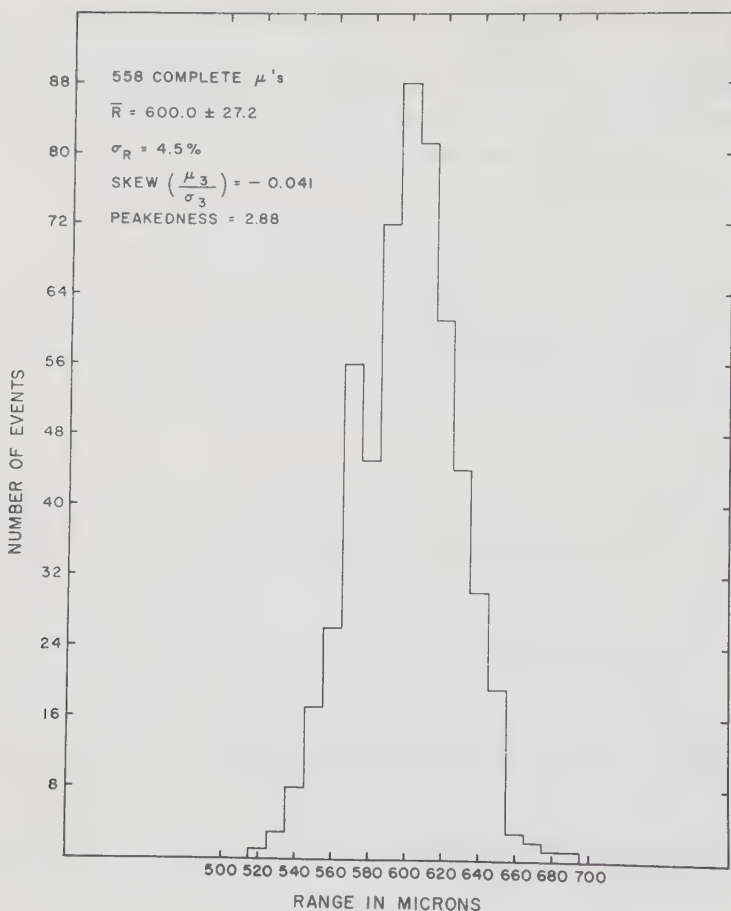


Figure 19. Observed range distribution of a mono-energetic group of positive muons having kinetic energy of 4.12 Mev.

We are now carrying out measurements on ions of  $C^{12}$ ,  $N^{14}$ ,  $O^{16}$ ,  $Ne^{20}$ , and  $A^{40}$ . Figure 20 shows the appearance of the tracks of these ions in G.5 emulsion. In Figure 21 are shown 400 Mev tracks of  $A^{40}$  nuclei in various emulsions. It is obvious that primary ionization and track width are essentially unrelated quantities; the width being a function of delta ray range, emulsion sensitivity, and type of development. The primary ionization is best observed with insensitive emulsions. To illustrate the effect of development I now show some remarkable tracks of  $A^{40}$  in Figures 22 and 23. The tracks shown were first observed by Dr. Muga in a uranium-loaded emulsion; similar tracks are produced, we have found, merely by strong D-19 development.

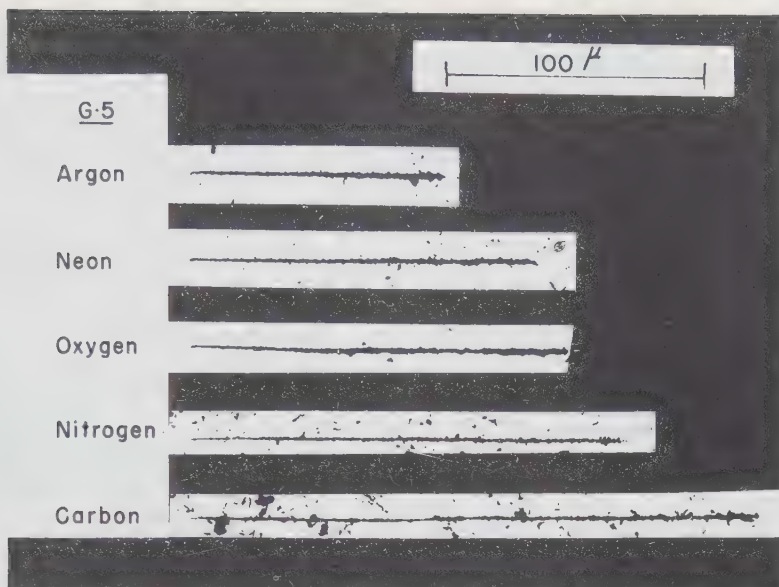


Figure 20. Photomicrographs of tracks produced by heavy ions of  $\sim 10$  Mev/nucleon in G.5 emulsion.

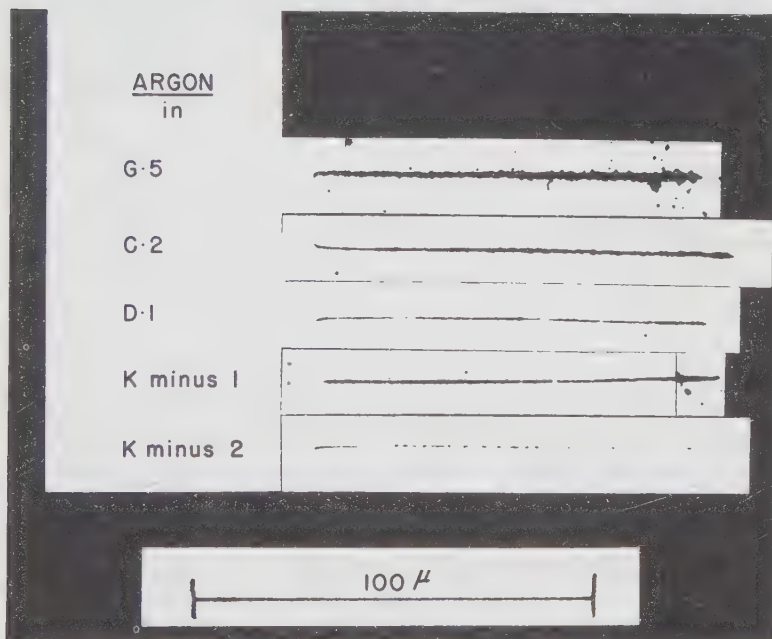


Figure 21. Photomicrographs of tracks produced by  $A^{40}$  ions of 400 Mev in various Ilford emulsions.



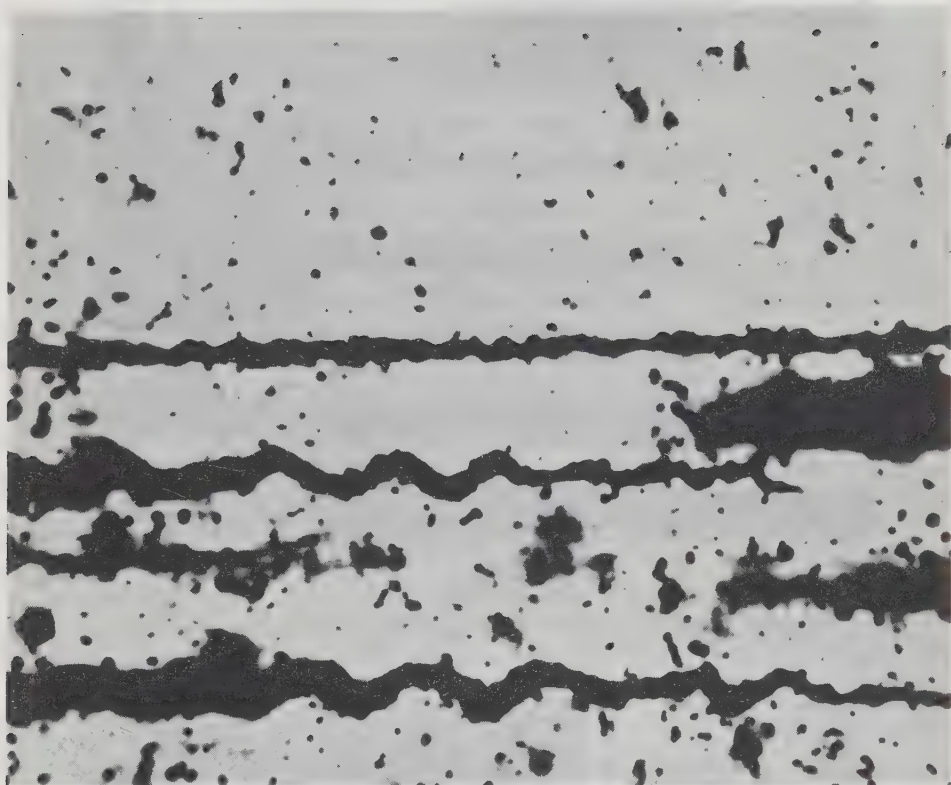


Figure 22. Tracks of  $A^{40}$  observed in Uranium-loaded Ilford C. 2 emulsion. The straight track is that of a nitrogen ion. The uranium loading appears to produce profound physical development of tracks in which the rate of energy loss exceeds 3 or 4 Mev/micron.

Although buckled tracks have been seen before occasionally, the connection of this effect with the ionization and development has not heretofore been fully realized. My tentative explanation is that, when the ionization is very intense, physical development causes neighboring grains to grow enough so that there is insufficient room for them all and the track must buckle. A unit of length--the buckling wave length--appears to exist.

Precision measurements for which Dr. H. H. Heckman should be given major credit are now being made on the ionization and range of heavy ions in emulsion.

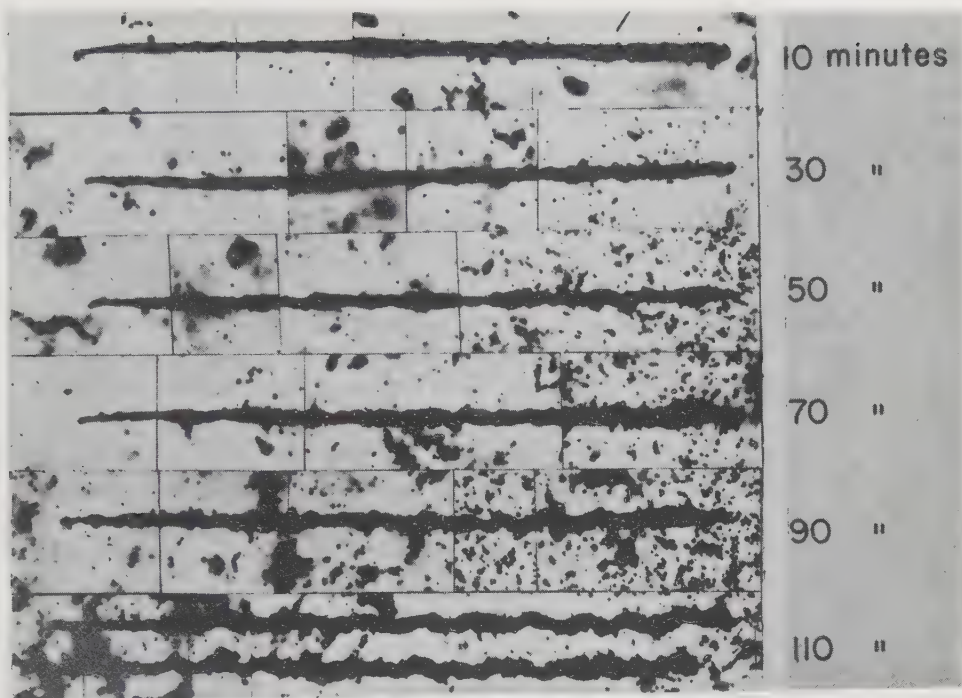


Figure 23. Photomicrographs demonstrating the development of buckling in tracks of  $A^{40}$  ions as the time of development with D19 developer is increased. Unloaded Ilford C.2 emulsion was the recording medium.

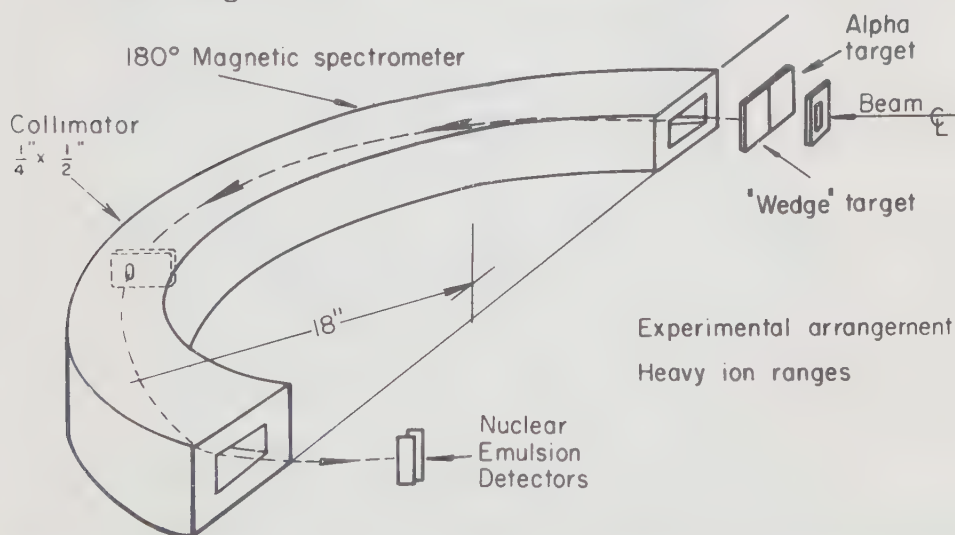


Figure 24. Schematic diagram of the magnetic spectrometer used for measuring heavy ion ranges.

We have employed the heavy ion linear accelerator to produce beams of various ions which we then study using a  $180^\circ$  magnetic spectrometer. This is illustrated in Figure 24. It gives us very high momentum resolution--high enough, in fact, so that we can also investigate the natural range straggling of even these heavy ions. We have employed this instrument in an advantageous way by the use of a wedge degrader shown in Figure 25. On admitting ions to the instrument through an absorber of graded thickness as shown, all ion velocities up to the maximum are present. Furthermore, all charge states are also represented, so that for each setting of the magnetic field intensity, several groups of ions having momenta that are integral multiples of the momentum of a proton passing through the same slit system are found on the plates. Without changing the field or moving the plate we also pass protons, alpha particles and other light nuclei through the system and record their tracks in the same emulsion plate. These serve as calibration tracks, giving us an over-all check of the magnetic field intensity and the emulsion density. This is also illustrated in Figure 25.

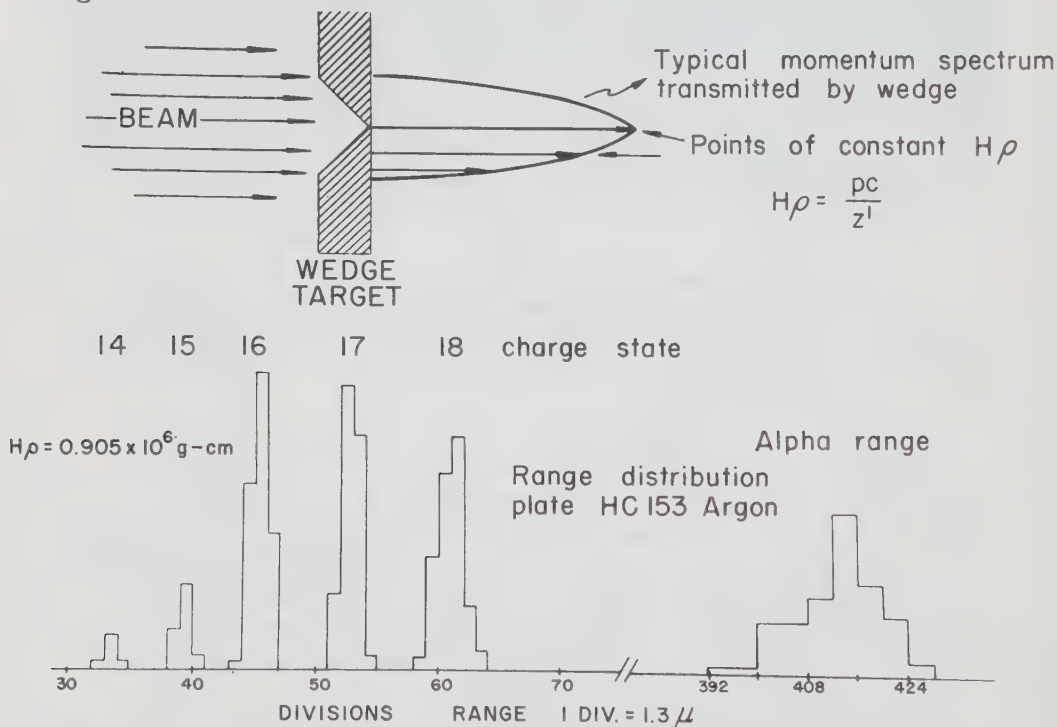


Figure 25. Illustration of how the variable thickness of target material produces ions of all velocities in all charge states. Subsequent magnetic analysis permits only those of a particular ratio of momentum to net charge to reach a detecting place.

At the lowest velocities we cannot resolve the tracks into their separate charge states, and are obliged to employ the collisions of the ions with protons of the emulsion to determine the initial form of the range-energy curve. The method is the same as that used by Miller in Berkeley several years ago.

The rather extensive series of measurements now under way are not yet completed, but our data, as they exist, are shown in Figures 26 and 27.

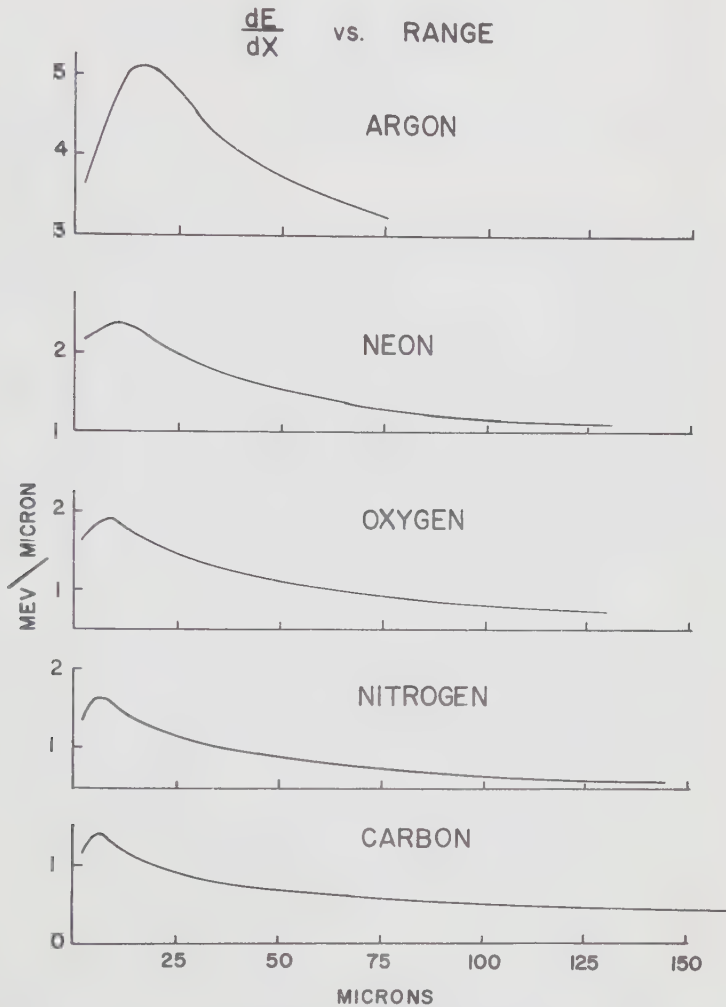


Figure 26. Energy loss rates in Mev/micron of  $C^{12}$ ,  $N^{14}$ ,  $O^{16}$ ,  $Ne^{20}$ , and  $A^{40}$  in standard emulsion.



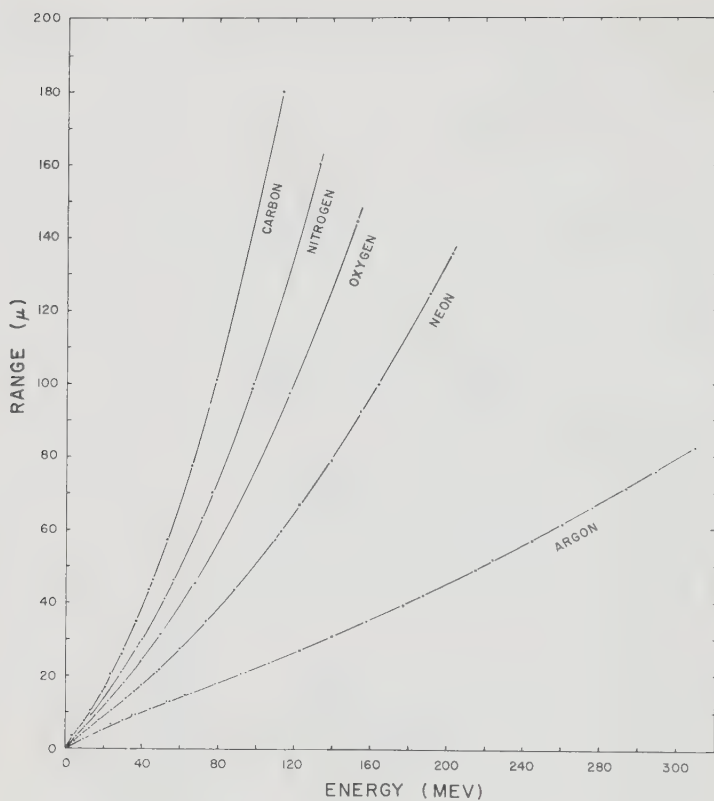


Figure 27. Ranges in microns of  $C^{12}$ ,  $N^{14}$ ,  $O^{16}$ ,  $Ne^{20}$ , and  $A^{40}$  in standard emulsion.

## II. RECENT DEVELOPMENTS IN THE THEORY OF STOPPING POWER

### 1. Principles of the Statistical Method

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The statistical method applied to the problems of stopping power can give comprehensive and rather accurate approximations. It is based on the assumptions that electron densities are high and that many electrons contribute significantly to the phenomenon in question, so that averages may be introduced. It points out similarities of different atoms. By studying it we may also take into account individual properties of atomic systems, retaining partially the statistical treatment. In the following we shall discuss some aspects of those stopping problems for which perturbation methods apply, corresponding to Bethe's treatment. The discussion is a continuation of a previous paper,<sup>33</sup> in the following referred to as A.

In the Thomas-Fermi atomic model the appropriate time scale is proportional to  $1/Z$ . This circumstance is essentially what one needs in order to derive Bloch's relation  $I = I_0 Z$ , and to observe further that the variable  $x = v^2/v_0^2 Z$  may be useful when comparing the stopping powers of different elements. Thus, one may plot

$$L = \left( - \frac{de}{dr} \right) \frac{mv^2}{4\pi z^2 e^4 Z N}$$

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<sup>33</sup>J. Lindhard and M. Scharff, Kgl. Danske Vidensk. Selskab Mat.-Fys. Medd. 27, No. 15 (1953), in which paper was utilized results concerning stopping by a free electron gas derived in J. Lindhard, Kgl. Danske Vidensk. Selskab. Mat.-Fys. Medd. 28, No. 8 (1954).

against  $x$ , and in first approximation find a common curve for all elements, i. e.,  $L = L(x)$ . In plotting this curve no  $K$ ,  $L$ , ...-shell corrections should be made. In the previous paper, A, the empirical curve showed a considerable dip, which could not be fitted theoretically. Since that time numerous measurements have indicated a less pronounced effect, but considerable uncertainty seems to prevail, both as regards the  $I$ -values of the elements and as to the detailed behaviour of the stopping cross sections as functions of energy.

Starting from the statistical method as a first approximation, it is possible to treat individual differences between stopping curves. We shall study some examples in a rather simplified version, and shall be concerned primarily with the stopping of protons at quite low energies--down to and even beyond the maximum in stopping cross section--where departures from a Thomas-Fermi treatment should be particularly conspicuous. The investigation of such quite extreme cases by simple procedures can give us insight into the applicability of the statistical method and of the various approximations. At the same time it is hoped to evoke interest in further measurements at low proton energies.

For this purpose we assume that (cf. A, p. 24)

$$L = Z^{-1} \int_0^{2mv^2/\hbar C} g_Z(\omega) \cdot \log \frac{2mv^2}{\hbar \omega} d\omega \quad (1)$$

where  $C$  is a constant,  $g_Z(\omega)$  is the dipole oscillator-strength density of the substance in question, with  $\int_0^\infty g_Z(\omega) d\omega = Z$ . By using the simple cut-off procedure in (1) we have observed two requirements, viz., for high velocities  $L$  takes on the familiar asymptotic form, since the upper limit to the integral may be taken to be infinity, and moreover no atomic transition is permitted to give a negative contribution to  $L$  (if  $C \geq 1$ ). In the simple cut-off procedure  $C$  should be chosen somewhat larger than unity; a more accurate alternative to (1) would be to replace the logarithm in (1) by a smooth function of  $y = 2mv^2/\hbar \omega$ , behaving as  $\log y$  for large values of  $y$  varying as  $y^{3/2}$  for  $y$  tending to zero, and to let the integration go to infinity.

Consider next the dipole oscillator-strength density,  $g_Z(\omega)$ . In the Thomas-Fermi description it is a unique function of  $\omega/Z$ , which leads to the relation of Bloch,  $I = I_0 \cdot Z$ . In actual atomic systems the oscillator-strength density will deviate from the

Thomas-Fermi spectrum in various ways. First, in the low frequency end there is a strong reduction in the spectrum below a frequency of order of the Rydberg frequency, and in this region considerable fluctuations occur due to differences in binding of the outermost electrons. This behaviour of the spectrum will cause a gradual rise of the average  $I/Z$  with decreasing  $Z$  and individual fluctuations in  $I/Z$ . Second, for intermediate values of  $\omega/Z$  the atomic shell effects will give rise to fluctuations about the average spectrum. Third, at high frequencies of the order of  $Z^2$  Rydberg,  $g_Z(\omega)$  must deviate from the Thomas-Fermi spectrum, but the oscillator strength of this region is rather small. Now, it seems reasonable to suppose that the deviations in the intermediate and high frequency part of the spectrum may be neglected in first approximation and, therefore, that effects due to the low frequency part dominate.

From expressions like (1) may be estimated not merely values of  $I/Z$ , but also the deviations of  $L$  from the curve  $\log(2mv^2/I)$ . For instance, if the Thomas-Fermi spectrum is used for  $g_Z(\omega)$  one may attempt to compute by the statistical method the equivalent of  $K$ ,  $L$ ,  $M$ , ...-shell corrections. For the present we shall be concerned with extremely low energies and use the crude approximation that in (1)  $C = 1$ . This can give a slightly distorted picture at medium energies but remains acceptable at low energies if the proper value of  $C$  is not large compared to 2.

In the absence of more accurate statistical estimates of  $g_Z(\omega)$  we can, as a guide, use the same approach as in A, based on collective effects. We assume that at a point of density  $\rho$  the effective frequency is

$$\omega = X \omega_0 = X(4\pi e^2 \rho/m)^{1/2} \quad (2)$$

putting, for definiteness, the constant  $X$  equal to  $\sqrt{2}$ . From a knowledge of  $\rho$  we can derive  $g_Z(\omega)$  on the basis of (2). since the oscillator strength ascribed to a volume element is assumed to equal the number of electrons in it, i. e.,  $4\pi \rho r^2 dr = g_Z(\omega) d\omega$ .

From (1) and (2) stopping curves were computed using atomic Hartree charge densities for Ne, Ar, Kr, Xe, O, N, Be, Al, Cu, and Au; for the metals a uniform distribution in space of valence electrons was assumed. In this way we first computed values of  $I/Z$  as listed in Table IV and then stopping-power curves for comparison with the compilations of W. Whaling.<sup>34</sup>

<sup>34</sup>W. Whaling, Hdb. der Physik XXXIV, 193 (1958)

TABLE IV

|         | N    | O    | Ne   | Al   | A    | Cu   | Kr  | Xe  | Au   |
|---------|------|------|------|------|------|------|-----|-----|------|
| I/Z(eV) | 13.4 | 13.6 | 14.7 | 13.0 | 10.9 | 12.6 | 9.9 | 9.7 | 10.2 |

As an example, Figure 28 shows the smoothed empirical curves of Whaling (solid line curves) for Ne, A, Kr and Xe, together with the present computations (dotted curves). The computed curves are not far in error even down beyond the maxima in the stopping cross sections. They exhibit oscillations of small amplitude, due to the atomic shells; the oscillations will be reduced somewhat if one does not use the sharp cutoff in equation (1).

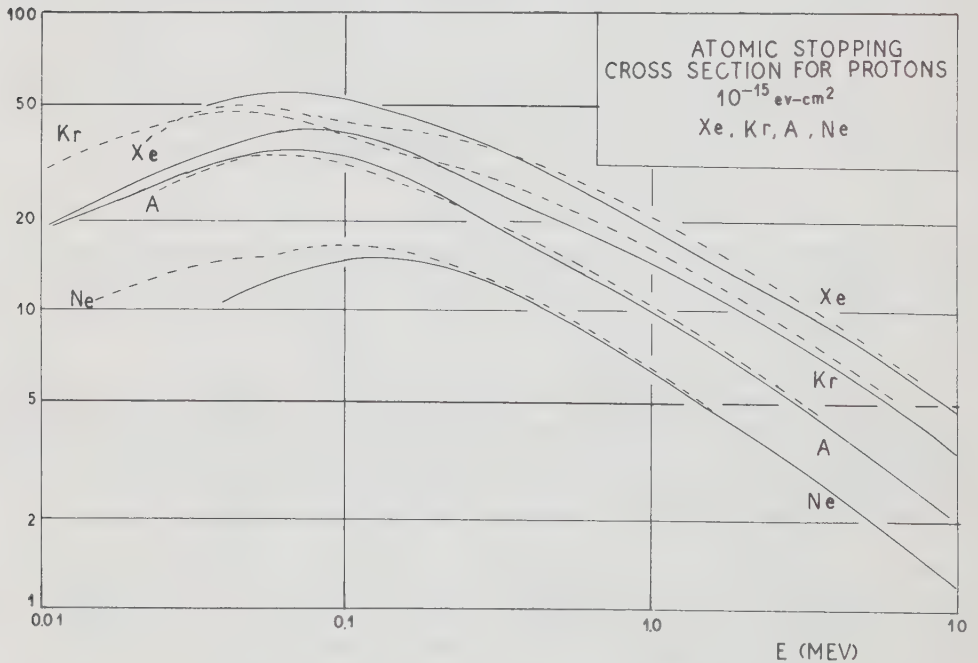


Figure 28. Comparison of empirical and theoretical curves for stopping cross sections in the noble gases. The solid line curves are the smoothed experimental curves; the dotted line curves are calculated according to the procedure described in the text.



These computations have perhaps only a qualitative significance, but they indicate that many electrons contribute to the energy loss even at low energies. Moreover, deviations in stopping power from the Thomas-Fermi results seem to arise mainly from the low frequency part of the spectrum. We can therefore attempt to take up the stopping at low proton energies in a different manner, and try to extract the I-value of any substance from the experimental data at low energies. In this region conspicuous characteristics of the stopping are the position and the height of the maximum stopping cross section. We shall relate these quantities using the statistical description.

We introduce a normalized stopping cross section,  $s$ ,

$$s = L/x \quad , \quad \text{or} \quad \frac{dE}{dR} = 4\pi z^2 e^2 a_0 N \cdot s \quad (3)$$

According to (1), the oscillator sum contributing to the stopping is, for a given particle velocity,  $v$ ,

$$Z^* = \int_0^{2mv^2/\hbar} g_Z(\omega) d\omega \quad (4)$$

where we have assumed that  $C = 1$ . The maximum in the stopping cross section is determined by  $\partial s / \partial x = 0$ , and we denote the corresponding values of  $L$ ,  $s$ , and  $x$  by  $L_m$ ,  $s_m$ , and  $x_m$ . We then deduce from (1), (3), and (4) that at the maximum

$$L_m = s_m \cdot x_m = \frac{Z^*}{Z} \quad (5)$$

The quantities  $s_m$  and  $x_m$  may be measured, and from (5) we thus obtain the oscillator sum contributing to the stopping,  $Z^*$ . The average of  $\log \omega$  for this part of the spectrum is according to (1) determined by  $L_m$ . The remaining oscillator sum,  $Z - Z^*$ , we now assume is distributed as the high frequency part of the spectrum of a statistical model, e. g., the Thomas-Fermi model with frequency distribution according to (2). With this assumption the spectrum is sufficiently determined to permit an estimate of  $I/Z$  for the substance in question. In Table V is shown the resulting  $I/Z$  values for a number of substances, as derived by this semi-empirical method from the maxima in the curves compiled by Whaling. Evidently, the treatment may be improved upon, but it is interesting to observe that the  $I/Z$  values are not unreasonable and that there is a correlation to the values in Table IV, computed on a

TABLE V

|         | N <sub>2</sub> | O <sub>2</sub> | Ne   | Al   | A    | Cu   | Kr   | Ag   | Xe  | Au  |
|---------|----------------|----------------|------|------|------|------|------|------|-----|-----|
| I/Z(eV) | 14.3           | 14.8           | 16.2 | 13.0 | 10.2 | 12.2 | 10.0 | 10.4 | 9.7 | 9.9 |

purely theoretical basis from the density distribution of Hartree models. While the values of  $I/Z$  in Table IV are proportional to  $X$ , those in Table V are much less sensitive to  $X$ ; for the lower atomic numbers they are practically independent of  $X$ , and for high atomic numbers they behave as  $X^{1/2}$ .

At first sight the positions and heights of the maximum cross section for different substances appear to be distributed rather arbitrarily. The above treatment suggests correlations with the values of  $I/Z$ , and applicability of a statistical description also at low proton energies. Let us finally demonstrate the latter in a more cursory manner. We consider again equation (4) and note that in a simple statistical model  $g_Z(\omega)$  is proportional to  $(Z/\omega)^{1/2}$ . [Cf. A, eq. (9)]. This implies that the right hand side of (5) becomes proportional to  $x_m^{1/2}$ , and accordingly one will expect that

$$s_m \cdot x_m^{1/2} \approx \text{const.}$$

In Table VI this quantity is shown for a number of substances. Compounds may be treated in a similar manner.

TABLE VI

|                 | N <sub>2</sub> | O <sub>2</sub> | Ne   | Al    | A     | Cu    | Kr    | Xe    | Au    | Pb    |
|-----------------|----------------|----------------|------|-------|-------|-------|-------|-------|-------|-------|
| $s_m$           | 1.89           | 1.79           | 1.54 | 2.04  | 3.60  | 2.55  | 4.19  | 5.66  | 4.05  | 4.61  |
| $x_m$           | 0.43           | 0.425          | 0.54 | 0.227 | 0.145 | 0.177 | 0.088 | 0.048 | 0.112 | 0.083 |
| $s_m x_m^{1/2}$ | 1.24           | 1.17           | 1.13 | 0.98  | 1.37  | 1.07  | 1.25  | 1.24  | 1.35  | 1.33  |

In the above we have studied stopping for protons of energies down to 50-100 kev, and we have supposed that at these low velocities one may neglect the opposing effects of capture and loss on the one hand, and screening by an electron bound to the proton on the other. For ions of higher nuclear charge at the same low velocities it will be important that the ions carry electrons.<sup>35</sup>

Clearly, the present approach, extending statistical methods to extreme cases, is merely tentative. Still, the results appear to confirm the consistency of the statistical type of approach.

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<sup>35</sup>A discussion of such cases is given in J. Lindhard and M. Scharff: "Quasi-Elastic Collisions between Ions and Atoms" (in press).

## 2. Dependence of Energy Loss on Valence States

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Du Pont Radiation Physics Laboratory

The dependence of stopping power of atoms on their molecular state was pointed out by W. Bothe,<sup>36</sup> who observed that the chemical binding should change the characteristic frequencies of the outer electrons, resulting in a changed rate of energy loss. This effect should be most noticeable in the light elements, being masked in the heavier elements by the more abundant inner electrons which are much less affected by chemical binding than the outer ones. It has only recently become possible to observe such effects in the experimental data. Data of great precision are required as the stopping power depends only logarithmically on the excitation levels of the stopping atoms.

One does not expect to find any fundamental deficiencies in the stopping power theories. Rather, the hope is that the more accurate measurements, combined with the theory, will allow one to interpret and interrelate the stopping power with other independently measured properties of the stopping materials.

In this connection it is fortunate that a new source of experimental information has become available, namely the characteristic energy losses of charged particles in single collisions. This has led to what might be called a new spectroscopy, the spectroscopy of longitudinal electronic excitations in matter. This contrasts with the usual spectroscopy in that the excitations contain components due to the propagation of polarization waves in which the electronic motion is parallel to the polarization wave vector, whereas the usual optical spectra entail excitations in which the electrons are coupled to the transverse electromagnetic field.

There has been some reluctance to accept the results of the theory of longitudinal excitations in dense matter as a guide for interpreting energy losses. The reason perhaps lies in an over-emphasis on the successes of the plasma theory. The characteristic

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<sup>36</sup>W. Bothe, *Jahrb. d. Radioabst.* 20, 73 (1923).

energy losses and zero-energy polarization effects do also occur in molecular insulators to which the plasma theory does not apply.

The theory of longitudinal coupled field and material oscillators constitutes a careful examination of the virtual oscillators of the stopping power theory which are not particularly specified by the latter theory. For low densities of atoms and for tightly bound electrons, the Coulomb interaction between the excited electrons is weak and the longitudinal and transverse oscillators coincide. In dense media and for loosely bound electrons the excitation levels shift upwards due to the strong Coulomb interaction. This leads to the "zero-energy polarization" effect. The Bethe stopping power formula may be derived directly in terms of longitudinal excitation levels if one identifies the absorbing oscillators with those excited by the longitudinal field. This is true at non-relativistic speeds where electrostatic effects dominate. At high speeds the stopping is determined mainly by the electromagnetic interaction and, as expected, the stopping power levels off in the Fermi plateau. The probability of longitudinal excitation does not depend on speed. Since the material-characteristic effects are the subject of interest, the energies of the particles will be taken as moderate, say in the range 1 to 1000 Mev.

In this range of energy the stopping power may be written as

$$-dE/dR = (4\pi e^2(ez)^2NZ/mv^2) \left[ \log 2mv^2 - \log I_0 - \log f_{val} - \log f_i - \log f_\mu - \Delta_{pol} \right]$$

$\Delta_{pol}$  corrects for transverse polarization effects which are not discussed here.  $f_\mu$  gives the relativistic corrections which are dependent on the nature of the particle. The  $f_i$  gives the inner shell corrections and  $f_{val}$  the effects of the valence state.  $I_0$  is the mean excitation potential for an isolated atom. Experiments give results on  $I_{eff} = I_0 f_{val} f_i$  and one would like to extract  $I_0$  since this is the number of primary interest. Of considerable interest also are the factors  $f_{val}$  as a function of atomic binding, and  $f_i$  as a function of  $Z$  and  $E$ .

Burkig and Mackenzie<sup>6, 8</sup> have made the most detailed study of the  $Z$ -dependence of the stopping powers, measuring the stopping power relative to aluminum for 23 elements. In Figure 29 the ratio of mean excitation potential to atomic stopping number  $I/Z$  derived from the experimental data without corrections is plotted against atomic number  $Z$ . There are clear indications of fine structure in



## MEAN EXCITATION POTENTIAL COMPARED TO ELECTRON DENSITY

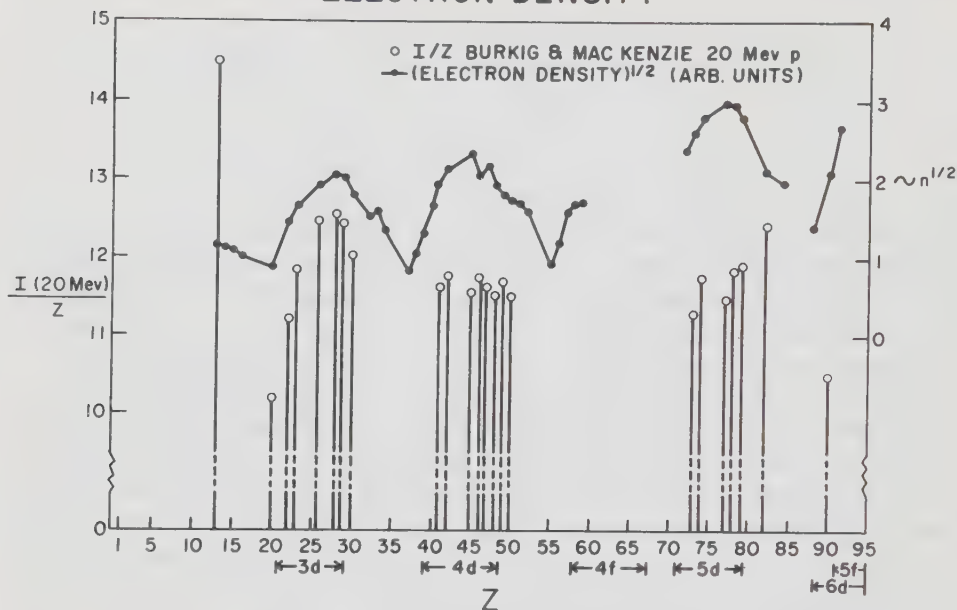


Figure 29. Shell effects in the mean excitation potential and electron density.

the region where the 3d shell is being filled. For the 4d and 5d shells the evidence is less conclusive, some of the critical elements being in doubt. The foils for Mo (42) and Rh (45) were not very uniform and hence these results are unreliable. There is an indication of a drop at Th(90) but the value for Pb (82) seems too high for the general trend. Figure 29 also contains a curve of the square root of the electron density in metallic solids which is proportional to the mean collective excitation frequency. The correlation with the variations in  $I/Z$  is quite good.

Valence state effects should show up as deviations from Bragg's empirical rule which states that the stopping power of compounds is equal to the sum of the stopping powers of the constituent atoms. One may take as a reference substance for comparison purposes the "Bragg-gas," a gas for which the Bragg rule holds precisely. This might be a very dilute monatomic gas.

Deviations from the Bragg rule arise from chemical binding, intermolecular interaction, and polarization. Figure 30 shows the

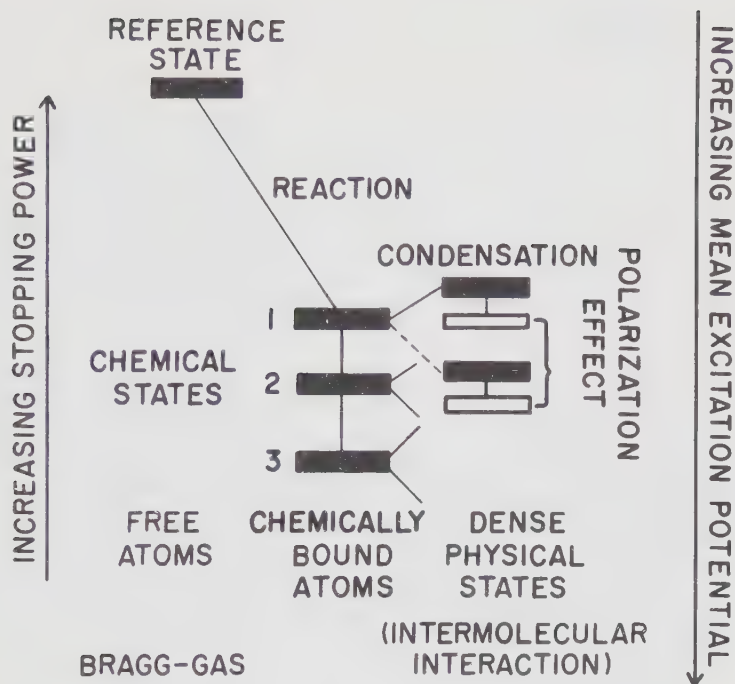


Figure 30. Valence and condensation effects on stopping power and the mean excitation potential.

relative positions with respect to stopping power and mean excitation potential of the various states of a substance. After a chemical reaction, an atom is in a state lower than the ground state of the free atom due to exchange forces which provide the bond. Thus the mean excitation potential is increased. The differences in  $I$  should have the same sign as the difference in bond energies per binding electron for different valence states but may be of larger magnitude.

There is no direct information as to the sign and magnitude of effects arising from intermolecular interactions. Finally the polarization effects which depend on the density of the material and its dielectric properties must be taken into account. The shielding reduces the energy transfer to the atoms and hence reduces the stopping power. It should be noted that frequently a shift from valence state 1 to a valence state 2, with  $I_1 < I_2$ , is balanced by a simultaneous decrease of polarizability. This partial compensation results in better apparent adherence to the Bragg rule than is to be expected from the change of valence states alone. Carbon in aromatic and aliphatic hydrocarbons has this behavior. On the other

hand, for the hydrogen in these compounds, the polarization effects appear to enhance the valence-state effects.

Figure 31 presents a summary of the valence effects in the lightest elements. The largest collective effects appear to be in LiH where  $I$  approximately doubles. Figure 32 shows the ratio  $f_{\text{val}} = I_{\text{val}}/I_0$  as a function of  $Z$ . As A. Bohr has pointed out, the ratio is unusually large for Be. It should be noted that also the value for Al is large, being nearly that of Be, and hence Al should not provide a good measure of the Bloch constant for heavier elements.

If  $f_{\text{val}}$  is known, values of  $I_0$  can be obtained from the experimental results. These values of  $I_0$  may then be compared with the predictions of the statistical model, that the ratio  $I_0/Z$  should be essentially constant. If the exchange interaction is included, the result becomes  $I_0/Z = K_0 (1 + k_0 Z^{-2/3})$ . Figure 33 shows the effect of applying the corrections implied in  $f_{\text{val}}$ . This correction accounts for the fluctuations of the uncorrected values. The corrected values are in good agreement with the form of the statistical model prediction with exchange.

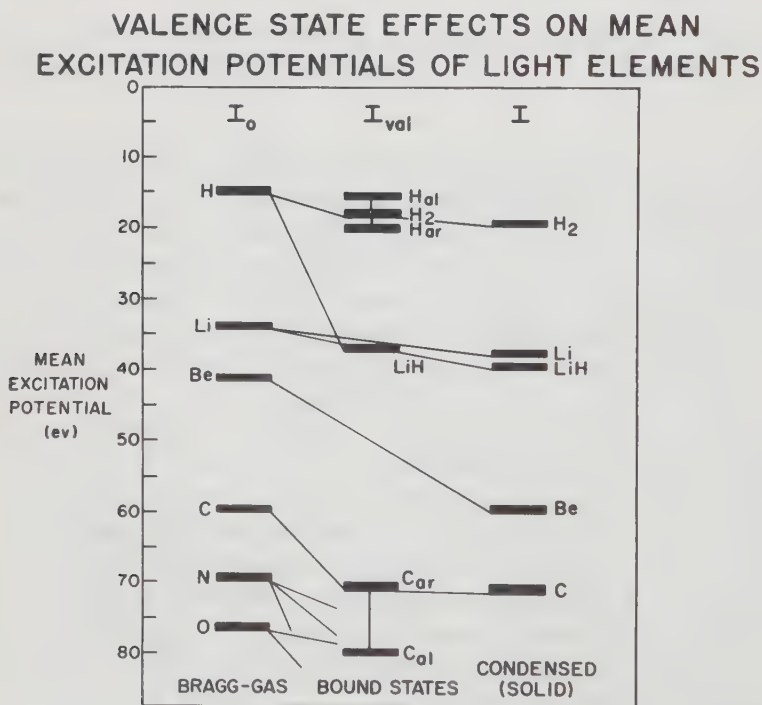


Figure 31. Valence and condensation effects on the mean excitation potentials in the light elements.

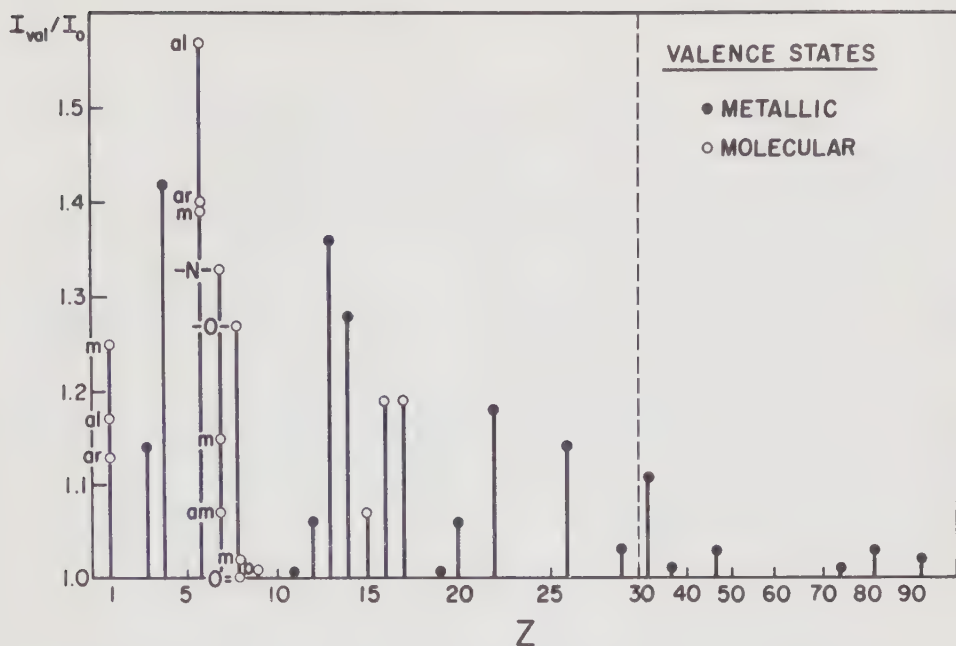


Figure 32. Valence corrections  $f_{\text{val}} = I_{\text{val}} / I_0$ .

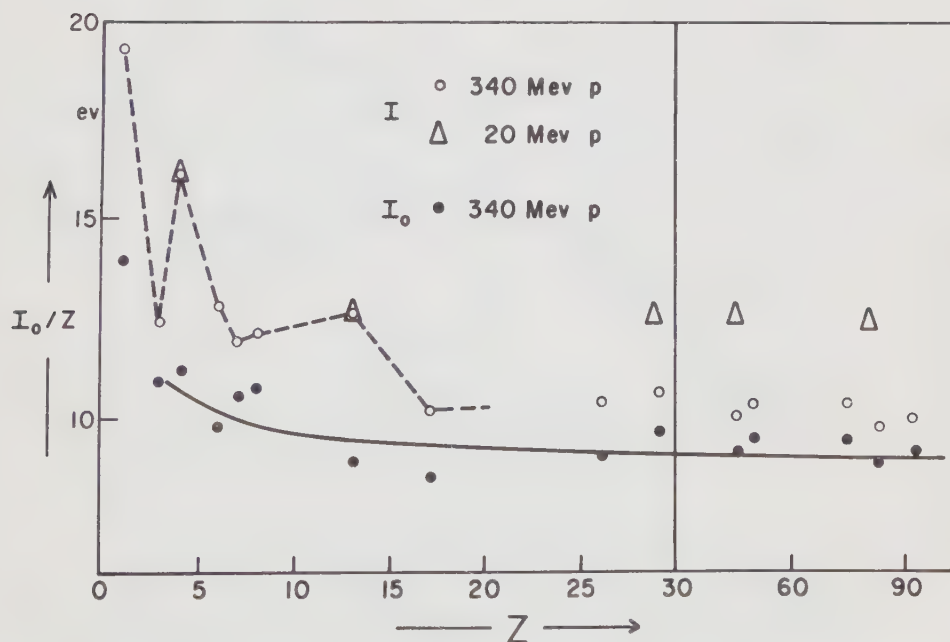


Figure 33. Ratio of the mean excitation potential  $I_0$  to the atomic number  $Z$ . The dashed line and open points indicate experimental values with no corrections. Solid points indicate values including corrections. The solid curve is that given by the statistical model including exchange.

It is difficult to interpret low energy data to see whether the exchange interaction form is correct. Walske<sup>19</sup> has calculated K and L shell corrections based on hydrogenic wave functions. Their extension to higher shells would be of dubious validity. Hartree type calculations would be reasonable in principle but of formidable computational difficulty. The statistical model ought to provide an estimate of stopping power deficiency of electrons in the bulk of the atom. A calculation has been made with a simple cutoff model in which electrons of frequency  $\omega_0$  do not contribute if  $\log 2mv^2/\hbar \omega_0 < 0$ . Figure 34 shows the shell correction factors  $f_i$ . The two theoretical inner-shell corrections for 20 Mev protons agree well for  $Z$  below 73. Walske's inner shell corrections are good in the region  $Z$  less than 30 where only K and L shell effects can be observed.

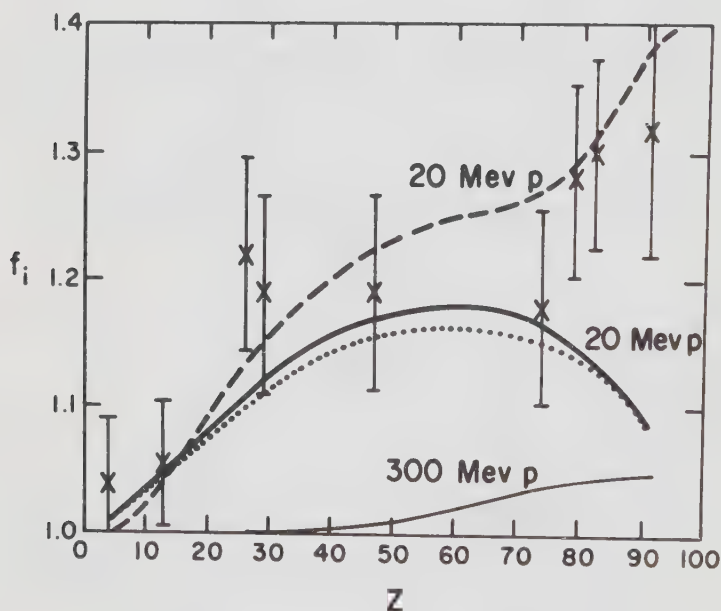


Figure 34. Tight binding corrections for 20 Mev protons. The dotted curve is Walske's K and L shell corrections, the solid curve the correction from the statistical model and the dashed curve the experimental result.



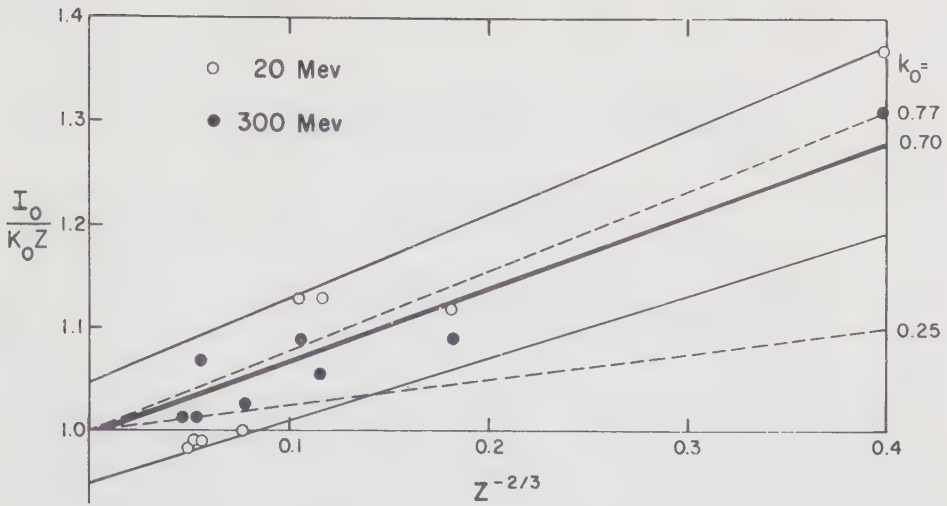


Figure 35.  $I_0/K_0Z$  versus  $Z^{-2/3}$ . The quality of fit to the statistical model with exchange for various values of  $k_0$  is represented by the various lines.

Using these corrections the discrepancies between high and low energy disappear as shown in Figure 35. The best value of  $K_0$  is found to be  $9.2 \pm 0.5$  ev. The value of  $k_0$  is less well determined, lying in the range 0.25 - 0.77. These numbers would be subject to slight changes when better scattering corrections are made to the existing data.

### III. CHARGE CHANGING COLLISIONS

#### 1. Introduction

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University of Chicago

Although the existence of collisions in which the charge of a moving gas ion is changed has been known since the first decade of this century, the first quantitative data were not obtained until about 1922 when Rutherford and Henderson observed such collisions for the  $\alpha$ -particles from radioactive substances. Since that time the experimental evidence has accumulated, at first slowly, but since 1948 at a greatly accelerated pace. Some fields of physics which are affected by charge changing collisions are:

- (1) The attempt to produce a controlled thermonuclear reaction, in which ions confined in a plasma retained in a magnetic field may escape by neutralizing themselves through electron capture.
- (2) The tandem Van de Graaff principle depends on charge changing collisions for its multiple acceleration from the same electrostatic potential.
- (3) The presence of Doppler shifted hydrogen lines in the aurora shows that incoming protons are capturing electrons in the atmosphere in collisions with gas molecules.
- (4) Some of the attempts to produce polarized beams of ions for acceleration, and subsequently for nuclear transformations, depend on charge changing collisions.
- (5) Attempts to solve theoretically the problem of capture of an electron by a moving positive ion have focused attention on some important but unclear aspects of collision theory when more than two particles are involved.

## 2. Electron Capture and Loss Cross Sections

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There are three known charge states of the hydrogen atom which are involved in the current electron capture and loss experiments, namely  $H^-$ ,  $H^0$ ,  $H^+$ . Using a currently accepted notation  $\sigma_{if}$  for a cross section,  $i$  being the initial, and  $f$  the final electronic charge of the projectile in the collision, we may say that in the limited kinetic energy range 10 to 30 kev, measurements of the six possible cross sections  $\sigma_{10}$ ,  $\sigma_{11}$ ,  $\sigma_{01}$ ,  $\sigma_{01}$ ,  $\sigma_{11}$ ,  $\sigma_{10}$  are available in the target gases  $H_2$ , He,  $N_2$ ,  $O_2$ , Ne, and A. The two cross sections  $\sigma_{01}$  and  $\sigma_{10}$  have been measured over the wide range from 0.2 to 1000 kev in  $H_2$ , He,  $N_2$  and A. Other scattered values, such as  $\sigma_{10}$  are known in these gases, and in  $CO_2$ .<sup>37</sup> The existing data on simultaneous loss of two electrons by  $H^-$  in the heavy noble gases shows some interesting anomalies which should be verified. Krypton seems to be less efficient as a target gas in causing such double losses than are the neighboring noble gases argon and xenon.

Equilibrium charge compositions in a hydrogen beam traversing gases can be computed from these measurements and, in many cases, have been directly measured. Where the measurements of different observers overlap, there is reasonably good agreement, the accuracy of the present techniques being something like 10 per cent. In particular the measurements of Kasner and Donahue (1957) on  $\sigma_{10}$  in hydrogen gas, from 2 to 60 kilovolts, have very recently been repeated, and the previously existing discrepancy between their values and those of Stier and Barnett has been removed.

In the case of moving helium atoms, four charge states have been reported in the literature. They are  $He^-$ ,  $He^0$ ,  $He^+$ ,  $He^{++}$ . Little is known of  $He^-$ , except its equilibrium ratio (see a later section of this report) and estimates of the probability of its

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<sup>37</sup>References to measurements discussed here will be found in a review article by Samuel K. Allison, Rev. Mod. Phys. 30, 1137 (1958).

formation from double electron capture by  $\text{He}^+$  in the noble gases in the kinetic energy range 60 to 175 kev. In the region 100 to 450 kilovolts kinetic energy, there are experimentally determined values of  $\sigma_{01}$ ,  $\sigma_{10}$ ,  $\sigma_{12}$ ,  $\sigma_{20}$ ,  $\sigma_{21}$  and an upper limit for  $\sigma_{02}$  in the gases  $\text{H}_2$ , He and air.  $\sigma_{01}$  and  $\sigma_{10}$  have been measured from 0.2 kev to 450 kev in  $\text{H}_2$ , He, and from 0.2 to 200 kev in  $\text{N}_2$ ,  $\text{O}_2$ , air, Ne, A, Kr, and Xe. There are serious discrepancies in the measurement of  $\sigma_{10}$  in krypton and xenon by various observers. In air there are scattered values of  $\sigma_{12}$  and  $\sigma_{21}$ , obtained from  $\alpha$ -particles from radioactive sources at energies from 646 to 6780 kev. Using  $F_i$  and  $F_{i\infty}$  to represent the fraction of electronic charge  $i$  under transient conditions or at charge equilibrium respectively we may state that direct measurements of the fractions  $F_{0\infty}$ ,  $F_{1\infty}$ ,  $F_{2\infty}$  in a charge equilibrated helium beam have been made from 4 to 480 kev in  $\text{H}_2$ , He, air, and A and from 4 to 200 kev in  $\text{N}_2$ ,  $\text{O}_2$ , and Ne.  $\text{He}^-$  will be mentioned later. In some instances, such as the region 200-480 kev, there is reason to believe that the equilibrium fractions computed from cross-section measurements are somewhat more accurate than those directly measured. In the energy region 480 to 6780 kev the equilibrium fractions of  $\text{He}^+$  and  $\text{He}^{++}$  in air have been computed from cross-section measurements.

The measurements of cross sections thus far reported might be referred to as total, in the sense that no analysis was made of the angular distribution of the products of the charge-changing collision, integration over all angles being included in the technique of the experiment itself. There is information on the angular distributions especially in the collisions of noble gas ions with noble gas targets at energies from 25 to 50 kev. These will be further detailed subsequently in this report.

For moving lithium ions and atoms, measurements of some charge-changing collision cross-section sums are available in the energy range 10-50 kev in the target gases  $\text{H}_2$  and  $\text{N}_2$ , with scattered measurements outside this range. Hasted<sup>38</sup> has made some observations on the detachment cross sections for  $\text{Li}^-$  ions between 0.025 and 4.0 kev in the target gas neon.

Measurements of the equilibrium ratios  $F_{1\infty}/F_{0\infty}$  and  $F_{2\infty}/F_{0\infty}$  for lithium ions of 20-50 kev kinetic energy in the gases  $\text{H}_2$ ,  $\text{N}_2$ ,  $\text{N}_2\text{O}$ , and  $\text{C}_3\text{H}_8$  have been made and will be mentioned later. The

<sup>38</sup>J. B. Hasted, Proc. Roy. Soc. Lond. A 212, 235 (1952).

equilibrium charge of a lithium ion beam emerging from a celluloid film at kinetic energies 500 to 5000 kev has recently been investigated.<sup>39</sup>

In some recent Russian work<sup>40</sup> ions of the elements boron, carbon, nitrogen, oxygen, and neon were accelerated in a 72 cm cyclotron to velocities in the range  $3.5 \times 10^8$  to  $8-11 \times 10^8$  cm/sec, in the case of nitrogen-14 corresponding to kinetic energies from 896 to 8860 kev. After passing beams of such ions through hydrogen, air, and argon as target gases in sufficient amounts to produce charge equilibration, the beams were analyzed for their charge components. A similar experiment was performed on such beams emerging at charge equilibrium from a celluloid film. The average ionic charge was 10 to 30 per cent larger after emerging from the celluloid than it was for gas equilibration. The distribution of charges of various amounts with respect to the average charge  $\bar{ze}$  was approximately Gaussian.

Double electron capture by  $C^+$  and  $O^+$  ions of kinetic energies in the range 10.7 to 54.5 kev has been reported.<sup>41</sup> Cross-sections  $\sigma_{11}$  have been measured in the target gases He, Ne, Ar, Kr, Xe,  $H_2$ ,  $N_2$ , and  $O_2$ .

In the case of moving  $C^+$  and  $O^+$  ions, measurements of single electron-capture probabilities  $\sigma_{10}$  in the energy range 0.2 to 4.0 kev have been reported by Hasted,<sup>42</sup> who also measured electron loss cross sections  $\sigma_{10}$  from negative oxygen ions in the same energy range. Charge distributions of 8.7 Mev oxygen ions in argon gas have been reported by Hubbard and Lauer.<sup>43</sup>

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<sup>39</sup>Ia. A. Jeplova, I. S. Dmitriev, V. S. Nikolaev, and L. W. Fateeva. J. Exptl. Theor. Phys. USSR 32, 974 (1957). Eng. Trans. JETP 5, 797 (1957).

<sup>40</sup>S. Nikolaev, I. S. Dmitriev, L. N. Fateeva, and Ia. A. Teplova, J. Exptl. Theoret. Phys. USSR 33, 1325 (1957). Eng. trans. JETP 6, 1019 (1958).

<sup>41</sup>Ia. M. Fogel, R. V. Mitin, and A. G. Koval. J. Exptl. Theor. Phys. USSR 31, 397 (1956). Eng. trans. JETP 4, 359 (1957).

<sup>42</sup>J. B. Hasted, Proc. Roy. Soc. Lond. A 212, 235 (1952); A 222, 74 (1954).

<sup>43</sup>E. C. Hubbard and E. J. Lauer, USAEC-UCRL-2778 (1954).



In the energy range 485 to 1180 kev, Korsunskii et al <sup>44</sup> have measured electron-loss cross sections for nitrogen atoms and also equilibrium charge distribution for nitrogen ions in gaseous nitrogen. Equilibrium charge fractions  $F_{0\infty}$  and the ratios  $F_{2\infty}/F_{1\infty}$  for 20-250 kev nitrogen ions in  $H_2$ , He,  $N_2$ ,  $O_2$ , Ne and A gaseous targets have been reported by Stier, Barnett, and Evans. <sup>45</sup>

Because of the ease with which 3 electrons can be stripped off the nitrogen atom,  $N^{3+}$  is frequently accelerated for heavy ion studies, and considerable information concerning the charge changing collisions of nitrogen ions in the million electron volt kinetic energy range is available. The charge composition at 15 Mev has been reported by Stephens and Walker, <sup>46</sup> and Reynolds, Wyly, and Zucker<sup>47</sup> have measured the nitrogen charge equilibrium as produced in formvar films in the kinetic energy range 6 to 26 Mev. Effective cross sections  $\sigma_{56}$ ,  $\sigma_{65}$ ,  $\sigma_{67}$ ,  $\sigma_{76}$  have been reported by the same authors for electron capture and loss of 26 Mev nitrogen ions in zapon. <sup>48</sup>

If we consider neon and the heavier noble gas ions as a group for discussion, we note that the charge distribution for 3.1, 7.1, and 10.3 Mev neon ions has been observed. <sup>49</sup> The ratios  $F_{2\infty}/F_{1\infty}$  and the equilibrium fraction  $F_{0\infty}$  for neon atoms and ions in the kinetic energy range 60-200 kev moving in the gases  $H_2$ , He,  $N_2$ ,  $O_2$ , Ne and A have also been measured. Similar studies have been

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<sup>44</sup>M. I. Korsunskii, L. I. Pivovar, A. M. Markus, and Kh. L. Leviant, Doklady Akad. Nauk USSR, 103, 399 (1955).

<sup>45</sup>P. M. Stier, C. F. Barnett, and G. E. Evans, Phys. Rev. 96, 973 (1954).

<sup>46</sup>K. G. Stephens and D. Walker, Phil. Mag. 45, 543 (1954).

<sup>47</sup>H. C. Reynolds, L. D. Wyly, and A. Zucker, Phys. Rev. 98, 474 (1955).

<sup>48</sup>H. C. Reynolds, L. D. Wyly, and A. Zucker, Phys. Rev. 98, 1825 (1955).

<sup>49</sup>Hubbard and Lauer, loc. cit.

published for moving argon atoms and ions.<sup>50</sup> The angular distribution of neon and argon ions scattered from charge changing collisions in noble gas targets in the energy range 25-100 kev has been studied by Everhart et al., and will be considered later in this report, together with similar studies by Russian observers.

Studies of electron detachment for heavy negative ions  $F^-$ ,  $S^-$ ,  $Cl^-$ ,  $Br^-$ ,  $I^-$ ,  $P^-$  moving through the noble gases have been reported<sup>51, 52</sup> in the kinetic energy range between 0.025 and 4.0 kev. Equilibrium ratios and single and double electron losses from the negative ions  $Cl^-$ ,  $Br^-$ ,  $I^-$ ,  $Na^-$ ,  $Sb^-$ ,  $Bi^-$ ,  $Bi_2^-$ , and  $Sb_2^-$  in the kinetic energy range 5-17.1 kev in the target gases argon and nitrogen have been observed by Dukelskii and Fedorenko.<sup>53</sup>

A study of the formation of negative ions of the heavy elements, such as Cu, Sb, Ag, Au, and Bi in the vapor phase, from various electron donor gases has been reported by Dukelskii.<sup>54</sup>

The extremely high energy, multiply-charged ions of the fission fragments offer opportunities for the study of single and multiple electron capture. Some such studies have appeared in papers from Copenhagen.<sup>55-59</sup> In this work some evidence was obtained

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<sup>50</sup>Stier and Barnett, loc. cit.

<sup>51</sup>Hasted 1952 loc. cit. and Proc. Roy. Soc. Lond. A 222, 74 (1954).

<sup>52</sup>V. F. Bidin and V. M. Dukelskii, J. Exptl. Theor. Phys. USSR 31, 569 (1956).

<sup>53</sup>V. M. Dukelskii and N. V. Fedorenko, Journ. Exptl. and Theor. Phys. USSR, 29, 473 (1955).

<sup>54</sup>V. M. Dukelskii, Doklady Akad. Nauk. USSR 105, 955 (1955).

<sup>55</sup>N. O. Lassen, Dan. Mat. Fys. Medd. 23, No. 2 (1945).

<sup>56</sup>N. O. Lassen, Phys. Rev. 68, 142 (1945).

<sup>57</sup>N. O. Lassen, Phys. Rev. 69, 137 (1946).

<sup>58</sup>N. O. Lassen, Phys. Rev. 79, 1016 (1950).

<sup>59</sup>Dan. Mat. Fys. Medd. 26, No. 5 (1951); 26, No. 12 (1951); and 30, No. 8 (1955).

that the electron loss cross section might be pressure dependent, and higher at higher pressures, where the time interval between successive collisions may be much shorter than the time to bind the disturbed electron again by radiating away electromagnetic energy. This matter has been discussed by Bohr and Lindhard<sup>60</sup> and by Bell.<sup>61</sup> A pressure effect on the cross sections has been occasionally suspected in experiments at lower energies with lighter moving ions, but it has not been definitely established. Closely associated with this question is the question of the purity of the beams of ions in common use for cross-section measurements. It has been the practice to assume that a beam of ions traversing the same orbit in an electric or magnetic field consists entirely of the same ionic species. Most investigators are well aware of the possibility of interference by particles of the same charge/mass ratio, familiar to the mass spectroscopists, but whether the electronic configuration of all the beam ions is the same, i.e., whether some are in metastable states, is a more subtle question and not so easily resolved. Barnett and Stier<sup>62</sup> found an apparent increase in the electron loss cross section of a neutral helium or neon beam if the beam is formed from  $\text{He}^+$  or  $\text{Ne}^+$  by electron capture at low rather than high gas pressures. They found no such effect in neutral hydrogen or nitrogen beams and concluded that, when formed at low pressures, the  $\text{He}^0$  and  $\text{Ne}^0$  beams contain significant fractions in the well-known metastable states of the noble atoms. There is, however, (and this was confirmed by Allison)<sup>63</sup> a wide region of pressure of the converter gas in which the electron-loss probability of the  $\text{He}^0$  beam is independent of the pressure at which it was formed. Whether there is a pressure-invariant fraction of metastable atoms when the neutral beam is formed in this pressure range remains unknown.

Another source of hidden errors in the present experimental techniques lies in the fact that most observations have been taken with apparatus in which the best obtainable vacuum was probably not much better than  $10^{-5}$  mm of mercury. The charge-changing cross sections are so large that in vessels of convenient length

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<sup>60</sup>N. Bohr and J. Lindhard, Dan. Mat. Fys. Medd. 28, No. 7 (1954).

<sup>61</sup>G. I. Bell, Phys. Rev. 90, 548 (1953).

<sup>62</sup>C. F. Barnett and P. M. Stier, Phys. Rev. 109, 385 (1958).

<sup>63</sup>S. K. Allison, Phys. Rev. 110, 670 (1958).

charge changing is still going on under "high vacuum" and must be corrected for. Although the most convenient and sensitive detector to use is probably the secondary electron-emitting surface, the question arises in measuring fractions of an ion beam bearing various charges whether two impinging ions, of equal kinetic energy but different electrical charge, will cause the emission of the same number of secondary electrons. Barnett and Reynolds<sup>64</sup> have examined this question for the impact of  $H^+$  and  $H^0$  ions at energies from 200 kev to 1000 kev on brass. The neutrals caused greater electron emission by a factor ranging from 1.37 at the lower energy to 1.62 at 1000 kev.

More work needs to be done on the double-electron captures. The measured values of the simultaneous pickup of two electrons by  $H^+$  to produce  $H^-$  seem surprisingly high. The method of formation of  $He^-$  from a  $He^+$  beam is unknown, and investigation might disclose some surprising intermediate steps.

If the recent experiments of Allison<sup>65</sup> are correct on the simultaneous capture of two electrons by  $He^{++}$ , this should be the predominate mode of neutralization of an  $He^{++}$  ion in gases such as air and helium at  $He^{++}$  kinetic energies somewhat below 100 kev.

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<sup>64</sup>C. F. Barnett and H. K. Reynolds, Phys. Rev. 109, 385 (1958).

<sup>65</sup>S. K. Allison, Phys. Rev. 109, 76 (1958).

### 3. Measurements Concerning the Negative Helium Ion

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It appears that the negative helium ion was first observed as a weak trace on a mass spectrogram by J. W. Hiby in 1939. In 1955 E. Holien and J. Midtdal calculated that the  $(1s\ 2s\ 2p)\ ^4P_{5/2}$  state of helium has an electron affinity of at least .075 ev. They attribute the formation of  $\text{He}^-$  as the attachment of an electron to an atom in the metastable  $(1s\ 2s)\ ^3S_1$  state.

With the aid of Chris Kuyatt and C. H. Sautter, studies have been made of the cross sections for formation and destruction of negative hydrogen ions, including some measurements on negative helium ions. For the helium experiments a beam of  $\text{He}^+$  ions is brought from the accelerator to a differentially pumped gas chamber 50 cm in length. Hydrogen is maintained in this chamber at various pressures from  $4 \times 10^{-3}$  mm to  $34 \times 10^{-3}$  mm. The beam emerging from this chamber is analyzed by catching the charged components on Faraday chambers. No measurements on the neutral helium component have been attempted.

In the work on hydrogen, the ratio  $\text{H}^-/\text{H}^+$  was measured as gas was admitted to the converter cell. The amount of  $\text{H}^-$  increased with increasing gas pressure and reached an equilibrium value in good agreement with those of Stier and Barnett.<sup>66</sup> When a similar experiment was performed with helium, the fraction  $\text{He}^-/\text{He}^+$  did not show a plateau, but it exhibited a maximum and then a falling off as the pressure was raised. Windham et al.<sup>67</sup> show a single curve of  $\text{He}^-$  beam plotted against hydrogen gas flow for 17.5 kev ions in their negative ion source. This curve shows a maximum but falls off much more rapidly than our curves do.

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<sup>66</sup>P. M. Stier and C. F. Barnett, Phys. Rev. 103, 896 (1956).

<sup>67</sup>P. M. Windham, P. J. Joseph, and J. A. Weinman, Phys. Rev. 109, 1193 (1958).



They interpret this in terms of a finding of Dukelskii et al.<sup>68</sup> that in Kr, Ar, and Ne gases helium negative ions are formed by double electron capture by positive ions. Their interpretation then involves the depletion of the  $\text{He}^+$  ions in the beam with the growth of the  $\text{He}^0$  component with the consequent decrease in the  $\text{He}^-$  beam. Since they do not give the gas thickness of their pickup tube, we cannot check the validity of this interpretation. However, we find that our curves obtained under more favorable conditions cannot be interpreted in this manner. Using cross sections obtained by Barnett and Stier, we find that equilibrium between the  $\text{He}^+$  and  $\text{He}^0$  is established at pressures lower than those at which the negative slopes occur.

As discussed previously in this summary Barnett and Stier have found a variation of cross section for electron loss for  $\text{He}^0$  and  $\text{Ne}^0$  with pressure of nitrogen in neutralizing chamber where their neutral beams are formed while no such variation occurs for  $\text{H}^0$  and  $\text{N}^0$ . They interpret this variation by stating that the character of the neutral beam for helium and neon was dependent on the pressure in the neutralizing chamber. Both helium and neon have metastable states and they assume a larger fraction of the beam is in the metastable states at low pressure than at high pressures.

The shape of our curves can be interpreted qualitatively in a similar manner. If we assume the  $\text{He}^-$  ions are formed from metastable neutral atoms as suggested by Holøien and Midtdal, and also that a beam of helium particles has its population of metastable atoms decrease with thickness of a gas target as suggested by Barnett and Stier, then the maximum in our  $\text{He}^-/\text{He}^+$  curves occurs where the metastable atom population is large, and this ratio decreases as this population decreases. We are unable to put this matter on a quantitative basis.

Table VII shows the values of the ratio  $\text{He}^-/\text{He}^+$  at the maxima of the growth curves for  $\text{He}^-/(\text{He}^0 + \text{He}^+ + \text{He}^{++})$ , obtained by using the information on  $\text{He}^0$  obtained from Barnett and Stier.<sup>69</sup>

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<sup>68</sup>V. M. Dukelskii, V. V. Afrosimov, and N. V. Fedorenko. Zhur. Eksptl. i. Teor. Fiz. USSR 30, 792 (1956). Eng. Trans. JETP 3, 764 (1956).

<sup>69</sup>C. F. Barnett and P. M. Stier, Phys. Rev. 109, 385 (1958).

TABLE VII

Maximum Amounts of  $\text{He}^-$  Formed by Charge Changing  
Collisions of  $\text{He}^+$  in  $\text{H}_2$  Gas

| $\text{He}^+$<br>Kinetic Energy<br>Kev | $\left\{ \frac{\text{He}^-}{\text{He}^+} \right\}_{\text{max}}$ | $\left\{ \frac{\text{He}^-}{\text{He}^0 + \text{He}^{++} + \text{He}^{++}} \right\}_{\text{max}}$ |
|----------------------------------------|-----------------------------------------------------------------|---------------------------------------------------------------------------------------------------|
| 40 Kev                                 | $2.97 \times 10^{-4}$                                           | $0.519 \times 10^{-4}$                                                                            |
| 50                                     | 4.12                                                            | 0.761                                                                                             |
| 60                                     | 5.31                                                            | 1.034                                                                                             |
| 80                                     | 6.70                                                            | 1.605                                                                                             |
| 90                                     | 7.06                                                            | 1.866                                                                                             |
| 100                                    | 7.03                                                            | 2.032                                                                                             |
| 110                                    | 6.84                                                            | 2.172                                                                                             |
| 120                                    | 6.65                                                            | 2.303                                                                                             |
| 130                                    | 6.17                                                            | 2.317                                                                                             |
| 140                                    | 5.75                                                            | 2.327                                                                                             |
| 160                                    | 4.85                                                            | 2.227                                                                                             |
| 180                                    | 3.92                                                            | 1.988                                                                                             |
| 200                                    | 3.14                                                            | 1.707                                                                                             |

#### 4. Some Measurements on the Formation of Negative Lithium Ions

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The ionization potential of  $\text{Li}^-$  has been calculated to be in the range 0.18-0.9 ev, <sup>70</sup> which is comparable to that for  $\text{H}^-$ , which is 0.75 ev.  $\text{Li}^-$  has been noticed by various observers and Hasted's measurements of some electron detachment cross sections for it have been previously mentioned in this review.

Some estimates of the velocity dependence of the lithium ion charge have been made by studying the density of ionization along the "handle" of "hammer" tracks in emulsions, <sup>71</sup> and a study of the equilibrium charge distribution involving  $\text{Li}^0$ ,  $\text{Li}^+$ ,  $\text{Li}^{++}$ ,  $\text{Li}^{+++}$  after passage of lithium ion beams through celluloid films has appeared. <sup>72</sup> The energy range used was 500 to 5000 kev.

The experiments presented here were done with a small accelerator in the energy range 10-50 kev.  $\text{Li}^+$  ions emitted from a hot filament coated with a lithium-aluminum silicate were passed through a converter cell 10.25 cm long, into which various gases could be introduced. The emergent beam from the cell was analyzed magnetically, the  $\text{Li}^-$  and  $\text{Li}^+$  being compared in a Faraday cup. For the ( $\sigma_{01} + \sigma_{02}$ ) cross section the attenuation of the  $\text{Li}^0$  beam was observed by relative readings of the secondary electron current ejected from a beryllium copper surface. An investigation of the increase of the  $\text{Li}^-/\text{Li}^+$  ratio as the converter cell pressure was increased did not show a maximum, as had been reported by Jorgensen

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<sup>70</sup>Ta-You Wu, Phil. Mag. 22, 837 (1936); Chin. J. Phys. 5 (1944); 6 (1945). E. Holþien, Arch. Mat. Naturvid, Oslo, LII 7 (1954).

<sup>71</sup>M. I. Kuznetsov, P. I. Lukirsky, and N. A. Perfilov, Doklady Acad. Nauk. USSR, 100, 665 (1955).

<sup>72</sup>Ia. A. Teplova, I. S. Dmitriev, V. S. Nikolaev, and L. N. Fateeva. Journ. Exptl. and Theor. Phys. USSR 32, 974 (1957). Eng. trans. JETP 5, 797 (1957).

for  $\text{He}^-$  production, but reached a plateau after about 4 microns of  $\text{N}_2$  had been introduced. Table VIII shows the observed  $F_{1\infty}/F_{1\infty}$  ratio in various gases.

TABLE VIII  
 $\text{Li}^-/\text{Li}^+$   
 Values of  $F_{1\infty}^-/F_{1\infty}^+ \times 10^4$  in Various Gases  
 Faraday Cell Detector

| Kinetic<br>Energy<br>Kev | $\text{H}_2$      | $\text{N}_2$    | $\text{N}_2\text{O}$ | $\text{C}_3\text{H}_8$ |
|--------------------------|-------------------|-----------------|----------------------|------------------------|
| 20                       | $0.28 \pm 0.03^*$ | $0.28 \pm 0.02$ | $1.0 \pm 0.1$        | $1.6 \pm 0.2$          |
| 30                       | $0.65 \pm 0.05$   | $0.87 \pm 0.06$ | $2.2 \pm 0.2$        | $3.8 \pm 0.2$          |
| 40                       | $0.97 \pm 0.05$   | $1.7 \pm 0.2$   | $3.6 \pm 0.3$        | $4.2 \pm 0.3$          |
| 50                       | $1.7 \pm 0.2$     | $1.9 \pm 0.3$   | $4.0 \pm 0.2$        | $3.8 \pm 0.3$          |

\*Extrapolation by secondary electron detector

An interesting feature of these results is the superiority of the more complex, lower ionization potential gases  $\text{N}_2\text{O}$  and  $\text{C}_3\text{H}_8$  (propane) for the production of  $\text{Li}^-$  at the lower kinetic energies, and the comparatively poor efficiency of  $\text{H}_2$  in this respect. An investigation at energies above 50 kev might well disclose, however, that in this region  $\text{H}_2$  is a good converter from  $\text{Li}^+$  to  $\text{Li}^-$ , as it is for  $\text{H}^+$  to  $\text{H}^-$  at about 15 kev kinetic energy.

The cross-section sums ( $\sigma_{01} + \sigma_{0\bar{1}} + \sigma_{02}$ ) for charge changing collisions of neutral beams, and ( $\sigma_{10} + \sigma_{1\bar{1}}$ ) for electron loss from the  $\text{Li}^-$  beam have been measured in the target gases  $\text{H}_2$  and  $\text{N}_2$  with the results shown in Table IX. It is reasonable to assume that at the velocities under investigation, double electron loss from neutral lithium is a negligible mode of charge change compared to single electron loss, so the cross section for the neutral beam is abbreviated to ( $\sigma_{01} + \sigma_{0\bar{1}}$ ).

The very large molecular cross sections for the removal of the outer electron in  $\text{Li}^-$  by nitrogen gas ( $290 \times 10^{-17} \text{ cm}^2$ , or 34 times the "standard" cross section  $\pi a_0^2$ ) illustrate the large size of the ion  $\text{Li}^-$  and the large impact parameters sufficient to transfer the requisite energy.

TABLE IX

Charge Changing Cross Sections for  $\text{Li}^-$  and  $\text{Li}^0$  Ions and Atoms  
 Cross Sections in Units of  $10^{-17} \text{ cm}^2$  per Target Atom of Gas

| $(\sigma_{\bar{1}0} + \sigma_{\bar{1}1})$ |              |              | $(\sigma_{01} + \sigma_{0\bar{1}})$ |              |              |
|-------------------------------------------|--------------|--------------|-------------------------------------|--------------|--------------|
| Target Gas                                |              |              | Target Gas                          |              |              |
| Kinetic Energy                            | $\text{H}_2$ | $\text{N}_2$ | Kinetic Energy                      | $\text{H}_2$ | $\text{N}_2$ |
| 10 kev                                    | 87           | -            | 10 kev                              | 15           | 26           |
| 20                                        | 100          | 120          | 20                                  | 17           | 37           |
| 30                                        | 92           | 130          | 30                                  | 16           | 31           |
| 40                                        | 84           | 145          | 40                                  | 12           | 29           |
| 50                                        | 76           | 118          | 50                                  | 10           | 23           |



## 5. The Spectroscopic Study of Electron-Capture Processes

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When high-energy charged particles are slowing down in the energy range for which charge changing collisions become one of the essential interactions with the stopping material, the energy transfer takes place in a far more complex form than when they are at high energies. Spectroscopic studies which reveal some of the interactions involved have been obtained as follows.

A beam of ions or electrons from an accelerator is directed by its magnetic analyzer into the collision chamber where the pressure of the target gas is maintained by means of differential pumping at 10 microns or less. The size of the collision chamber is 12 cm in diameter and 100 cm long. The light emission excited from the target gas is then photographed with a  $f/0.65$  grating spectrograph through windows in the side of the chamber.

Let us consider a well-known case as an example of electron transfer using  $N_2$  as the target gas. The ground state of  $N_2$  is singlet while the first three excited states are triplet. Therefore, the primary excitation of the triplet states which involves a flip of the spin of an electron can only be done by electron exchange. An examination of the spectra which have been excited by electrons of 23 and 500 ev and protons of 20 kev shows that the 1 PG and 2 PG are practically the only emissions in the spectrum excited by 23 ev electrons, but their intensity is negligible in the spectra excited by 500-ev electrons and 20-kev protons. This illustrates the role that the electron exchange plays in the energy transfer to the stopping material.

Let us now consider the energy loss through a charge-changing cycle. A positive ion  $A^+$  captures an electron from a target particle, then loses the electron by a subsequent collision with another target particle. Energy loss results from such a capture-loss cycle.

The electron can be captured into excited states as well as into the ground state. If the density of the stopping medium is low

enough that optical transition from the excited states can take place before "A" loses its electron, photons will be emitted. The emission is characterized by its Doppler shift when the spectrum is taken with the spectrograph aligned along the direction of motion of "A". Hydrogen lines with Doppler shifts can easily be seen in spectra in air at 10-micron pressure excited by 30-kev and 5-kev protons, respectively.

On the other hand, if the density of the medium is too high so that "A" loses its electron before the optical transition takes place, these lines disappear. Thus, the process of energy transfer in low density media is different from that in high density media. As a result, the value of the atomic stopping power  $-dE/dR$  becomes dependent on the density of the stopping medium, at least insofar as the losses in charge-changing cycles are an appreciable contribution to the total stopping power.

Let  $\sigma$  be the cross section of electron loss,  $N$  the particle density of the stopping medium, and  $v$  the velocity of the incident ion. The mean life of the captured electron is  $1/N\sigma v$ . If  $\tau$  is the lifetime of the excited state, then the density of the medium at which the contribution of the charge-changing cycle to the atomic stopping power is expected to be most sensitively density-dependent is determined by

$$\tau = 1/N\sigma v, \tag{1}$$

Take  $\sigma = 10^{-16} \text{ cm}^2$ ,  $v = 10^8 \text{ cm/sec}$ ,  $\tau = 10^{-8} \text{ sec}$ ; then  $N$  is  $10^{16}$  particles per cc, which is about 300 microns.

From the conservation of energy and momentum equations applied to the process of electron capture, one finds the energy transfer from the incident ion to the target particle is given by

$$E_2 = \frac{M_1 M_2}{M^2} \cdot E \left[ \cos \theta \pm \left\{ \cos^2 \theta - \frac{M}{M_2} \left( \frac{Q}{E} + \frac{m}{M_1} \right) \right\}^{1/2} \right]^2 \tag{2}$$

in which

- $m$  = electronic mass
- $M_1$  and  $M_2$  = respectively, the mass of the incident ion and of the target particle,  $M = M_1 + M_2 + m$
- $E$  = incident energy
- $\theta$  = recoiling angle of the target particle
- $Q$  = the difference between the binding energies of the electron in the two bound states.

Thus  $E_2$  is determined if  $\theta$  is known.

A cloud chamber picture of electron capture by helium ions in helium gas shows that large amounts of ions are ejected in the direction perpendicular to the direction of incidence, i. e., the angles of recoil are close to  $90^\circ$ . Accordingly, energy transfer for these processes is of the order of

$$E_2 = \frac{M_1}{M} E - \frac{Q}{E} + \frac{m}{M_1} \quad (3)$$

If we take a 25-kev proton and a 100-kev  $\text{He}^+$  ion capturing an electron from a nitrogen molecule to its ground state as examples, the values of  $E_2$  are 0.56 ev and 4.3 ev, respectively.

A spectroscopic study of such a process is possible only when the target particle is excited during the process of electron capture. The excitation of the negative system of  $\text{N}_2^+$  turns out to be such a case.

Let us consider the excitation of the 4709, 4652, 4600 Å bands by ionic impact. The upper vibrational quantum numbers of these bands are 0, 1, and 2, respectively. The reason for choosing these bands for study is that, in a low-dispersion spectrum these bands are close but still well separated, which greatly facilitates the comparison of their intensities. These bands can be excited from a nitrogen molecule either by direct ionization or by electron capture. It was found that at high energies their relative intensities are just the same as that excited by electronic impact, while at low energies the bands of higher vibrational quantum numbers are enhanced.

Comparing the excitation of these bands by 300 kev and 20 kev protons, respectively, we see that the three bands have noticeable but not pronounced intensity differences. But if we excite by 400 kev, 149 kev and 11 kev  $\text{He}^+$  ions, the bands of higher vibrational quantum numbers are greatly enhanced. This can be explained as the result of a forced vibration set in by the energy transfer during the process of electron capture. These variations seem to reflect exactly the increasing importance of electron capture at lower energies in ionizing the nitrogen molecules.

Equation (3) contains  $Q$ , the difference of the binding energies of the electrons in the two bound states. Since the value of  $Q$  for a molecule  $x_n y_m$  is in general different from that for  $x$  or  $y$ , it is expected that when the capture-loss cycle becomes one of the important processes of energy transfer, the additive rule for energy loss will no longer hold.

Analytically, the charge transfer process  $A_1^+ + A_2 \rightarrow A_1 + A_1 + A_2^+$  can be regarded as a net result of two interactions, the interaction between  $A_1^+$  and  $A_2$  which makes the reaction go to the positive direction and the interaction between  $A_1$  and  $A_2^+$  which makes the reaction go to the reverse direction. The energy transfer to  $A_2^+$  might be regarded as a measure of the strength of the interaction between  $A_1$  and  $A_2^+$ .

If this intuitive approach were correct, the charge transfer process would be more probable when the energy transfer is the least. It is seen from equation (2) that the energy transfer can never be zero except when the energy of the incident ion is equal to

$$E_0 = -\frac{M_1}{m} Q, \quad (5)$$

which is positive for an endothermic reaction. It happens that for all known cases the capture cross section reaches or almost reaches a maximum when  $E = E_0$ .

The additional maxima in the curves for  $H^+$  capturing in argon and neon can then be interpreted as the capture of inner shell electrons of argon and neon, respectively, by protons.

In conclusion, I would like to cite evidence showing the charge-changing process occurring in the auroral display. Auroral displays can be classified into two categories: quiet arcs and active rays.

Two sequence spectra have been taken of an aurora, the first of which was taken when it was an arc and the second was taken when it was rays. The intense hydrogen lines present in the first spectrum indicated the presence of protons. The absence of the hydrogen lines together with the high intensity of the first positive bands in the second spectra showed the role played by low-energy electrons in the display.

## 6. Coincidence Counting Methods of Studying Inelastic Ionic Collisions

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Table X shows various types of events in which a singly charged ion in collision with a neutral atom may itself lose electrons.

TABLE X

Notation for Collisions Involving Change of Charge

| Reference<br>Symbol | Process                                          | Descriptive<br>Name                                | Notation<br>for Cross<br>Section |
|---------------------|--------------------------------------------------|----------------------------------------------------|----------------------------------|
| (a)                 | $A^+ + B \rightarrow A^{m+} + B + (m-1)e$        | stripping                                          | $10^{\sigma_{mo}}$               |
| (b)                 | $A^+ + B \rightarrow A^+ + B^{n+} + ne$          | ionization                                         | $10^{\sigma_{ln}}$               |
| (ab)                | $A^+ + B \rightarrow A^{m+} + B^{n+} + (m+n-1)e$ | stripping and<br>multiple<br>ionization            | $10^{\sigma_{mn}}$               |
| (c)                 | $A^+ + B \rightarrow A + B^+$                    | charge<br>transfer                                 | $10^{\sigma_{01}}$               |
| (bc)                | $A^+ + B \rightarrow A + B^{n+} + (N-1)e$        | electron<br>capture plus<br>multiple<br>ionization | $10^{\sigma_{0n}}$               |

For complex atomic systems in the 100 kev region it is not yet possible to measure exactly specific collision cross sections either



by means of a double mass spectrometer (Fedorenko,<sup>73, 74</sup> Everhart<sup>75</sup>), or by the "condenser" method (Hasted<sup>76</sup>). In terms of the notations in Table X we may describe the physical quantities measured by various observers in the following way:

| Observers                        | Quantity Measured                                      | Designation                                             |
|----------------------------------|--------------------------------------------------------|---------------------------------------------------------|
| Everhart, Kaminker and Fedorenko | $\sum_{y=0}^m 10^{\sigma_{my}; m=1 \text{ to } 8}$     | (a + ab)<br>(stripping)                                 |
| Afrosimov and Fedorenko          | $\sum_{x=0}^m 10^{\sigma_{xn}; n=1 \text{ to } 4}$     | (b + c + bc)<br>charge trans-<br>fer plus<br>ionization |
| Hasted                           | $\sum_{y=0}^n 10^{\sigma_{Oy}}$                        | (c + bc)<br>charge<br>transfer                          |
|                                  | and                                                    |                                                         |
|                                  | $\sum_{x=1}^m \sum_{y=1}^n (m+n-1)_{10}^{\sigma_{xy}}$ | (a + b + ab)<br>stripping plus<br>ionization            |

<sup>73</sup>V. V. Afrosimov and N. V. Fedorenko, J. Tech. Phys. USSR, 26, 1872 (1957).

<sup>74</sup>D. M. Kaminker and N. V. Fedorenko, J. Tech. Phys. USSR, 25, 2239 1843 (1956).

<sup>75</sup>E. Everhart and co-workers, Phys. Rev., 99, 1287 (1955), 107, 704 (1957), 102, 1524 (1956).

<sup>76</sup>H. B. Gilbody and J. B. Hasted, Proc. Roy. Soc., A 238, 334 (1956), A 240, 382 (1957).

In the measurements of Hasted the degree of ionization was unknown. It is, of course, true that in many cases these measurements approximate to single cross sections.

Charge transfer collisions have a different type of energy dependence from those for ionization and for stripping, and although the theory of stripping developed by Russek and Tom Thomas<sup>77</sup> makes no distinction between reactions (a), (b), and (ab), it is unlikely that a theory could be developed to cover both (b) and (c); as yet, we know nothing about reaction (bc), which on the grounds of adiabatic theory alone would not be unlikely in the 100 kilovolt region for such collisions as  $\text{He}^+$  in A. We do not yet know if ionization reactions conform to the adiabatic theory, but we suspect that they do not, since their rate of rise with energy follows a different law. Charge transfer reactions conform to the equation

$$10^{\sigma_{01}} = A e^{-K_2 |\Delta E| / v}$$

as can be seen from Figures 36 and 37, which are obtained by analysis of the data of Hasted and other workers; but none of the ionization cross sections we have analyzed behave in this way. Figure 38 shows a typical ionization cross section, taken from low energy data for which multiple ionization can be ignored.

We do not know the energies of maximum cross section for ionization either from Hasted's or from Fedorenko's data. Although the agreement between the two is fairly good (Figures 39 and 40) it would undoubtedly be of value to have measurements of ionization and charge-transfer cross sections separately, together with information about the composite reaction (bc).

The only way this can be achieved is by coincidence counting of the products of the collisions. The principle of the experiment is shown in Figure 41. For charge-transfer collisions of types c and bc it is necessary to compare a count of coincidences between  $S_{n+P_0}$  with a count of  $P_+$ .

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<sup>77</sup>A. Russek and M. Tom Thomas, Phys. Rev., 109, 2015 (1958).

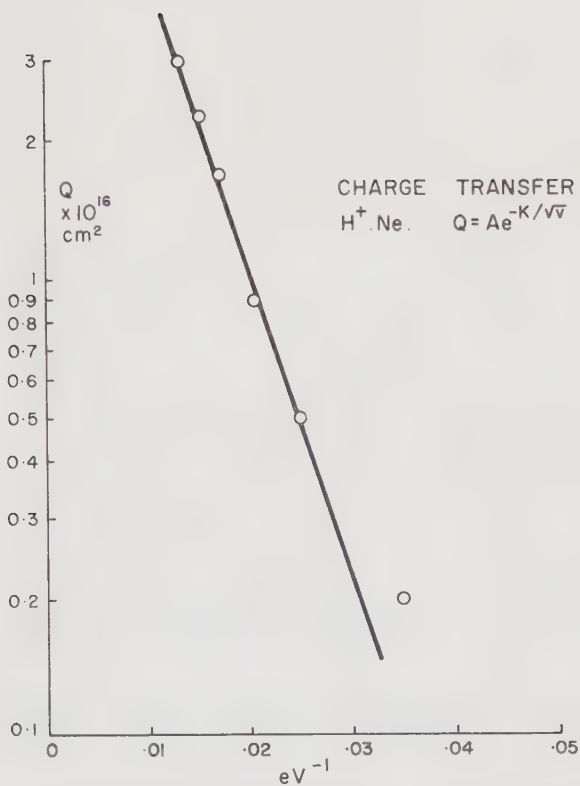


Figure 36. Analysis of charge transfer data for  $H^+$  in neon, conforming to the exponential law.

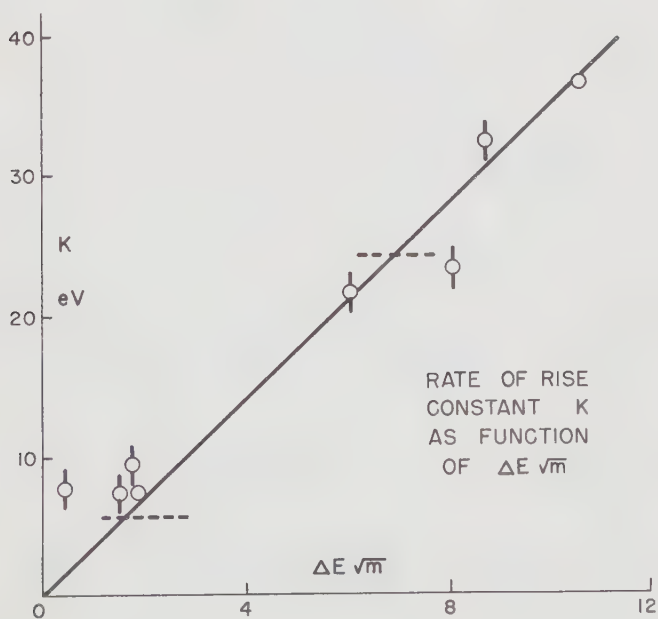


Figure 37. Rate-of-rise constant K as a function of  $\Delta E \sqrt{m}$ .

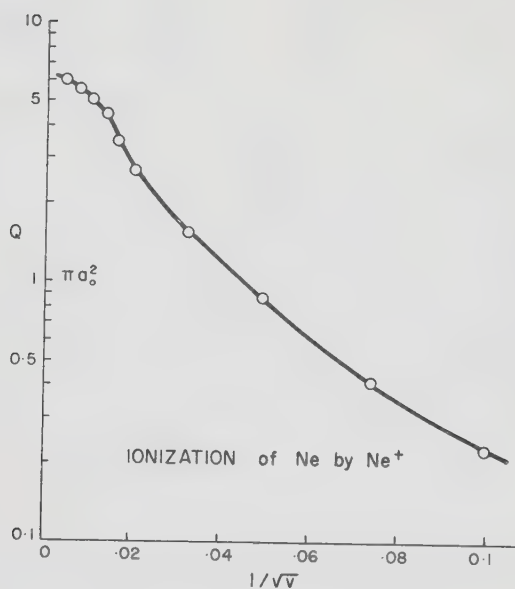


Figure 38. Analysis of the ionization of neon gas by  $\text{Ne}^+$  ions, which does not conform to the exponential law.

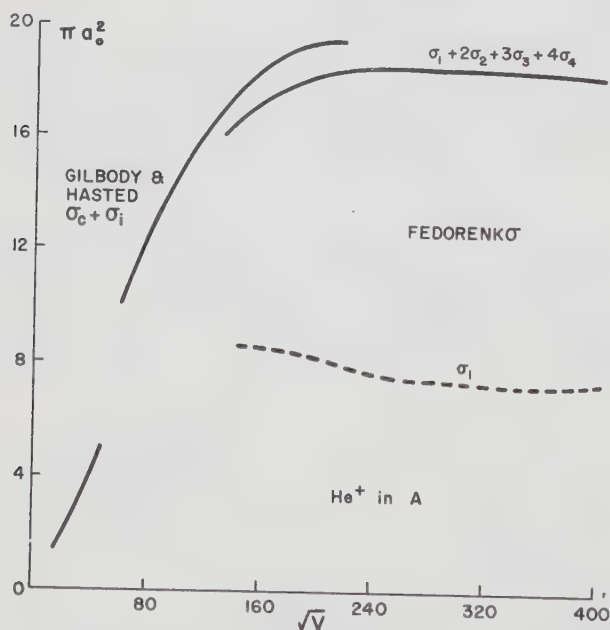


Figure 39. Russian and English data on ionization compared for  $\text{He}^+$  in argon gas.

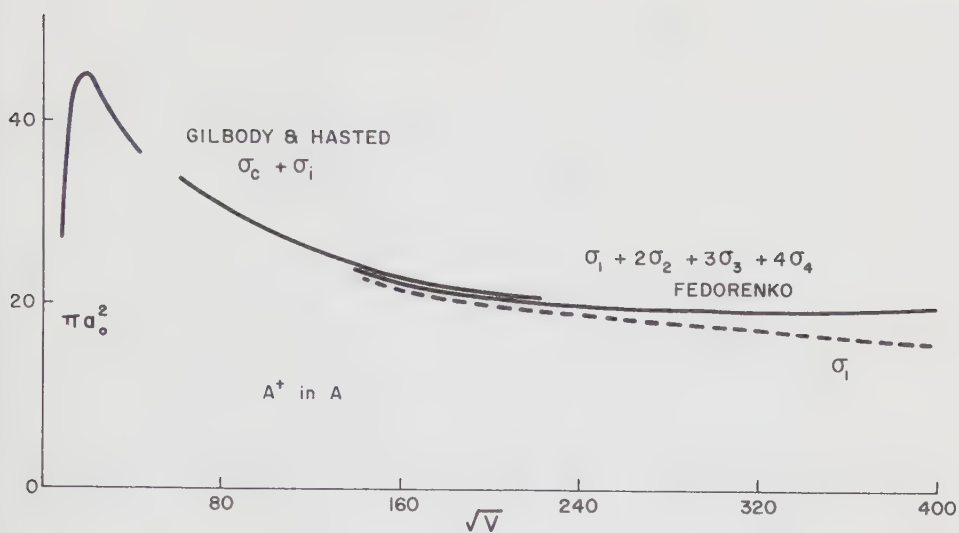


Figure 40. Russian and English data on ionization compared for  $A^+$  in argon gas.

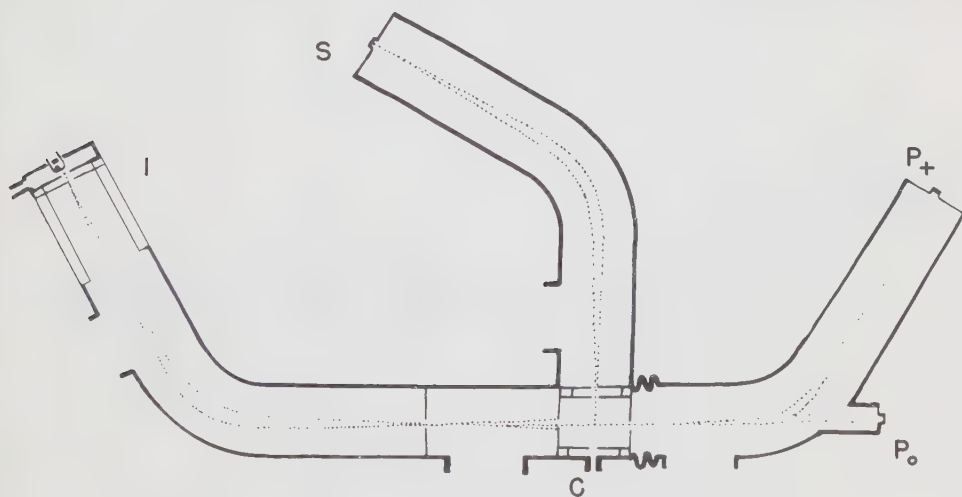


Figure 41. Schematic diagram of the coincidence counting experiment.



An apparatus for the range 3 - 40 kev, at present nearly completed, is shown in Figure 42. The mass analysis of the secondary beam is achieved by a  $60^\circ$  magnetic sector analysis, while the primary beam is analyzed through a small angle electrostatically. It is necessary that all the products of the collision for a given path length pass through the analyzer, and this has already been achieved for the secondary beam by Fedorenko, who widened his acceptance slits until no further increase in the beam current was observed. In the present apparatus the counting of single particles is attempted with a windowless EMI 9524 B multiplier with Sb-Cs dynodes, and photocathode removed. A fast coincidence unit of resolution  $10^{-8}$  second is used, with suitable adjustable delays to allow for the particle velocity. At a primary beam counting rate of  $10^5$  particles per second on an ERCO N530 scaler, this allows secondary beams of the order of 0.1 per cent of the primary to be distinguished in 10 seconds from a background of 100 counts per second. The layout of equipment using a non-pressurized van de Graaff generator for the range 40-500 kev is shown in Figure 43.

Numerous possible uses of such a technique will be apparent; in particular it should be of immediate importance to study the ratio of stripping to ionization cross sections.

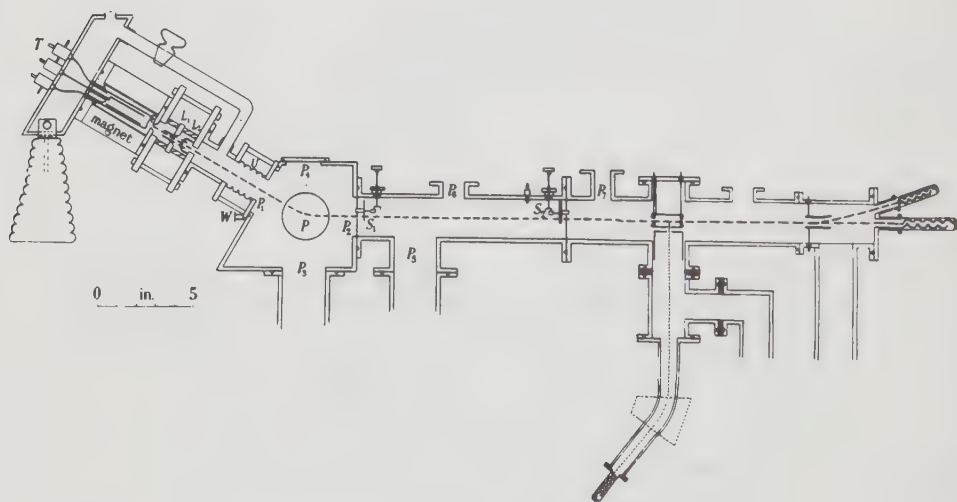


Figure 42. Double mass spectrometer, 3 - 40 kev.

500KEV VAN DE GRAFF  
POSITIVE ION ACCELERATOR

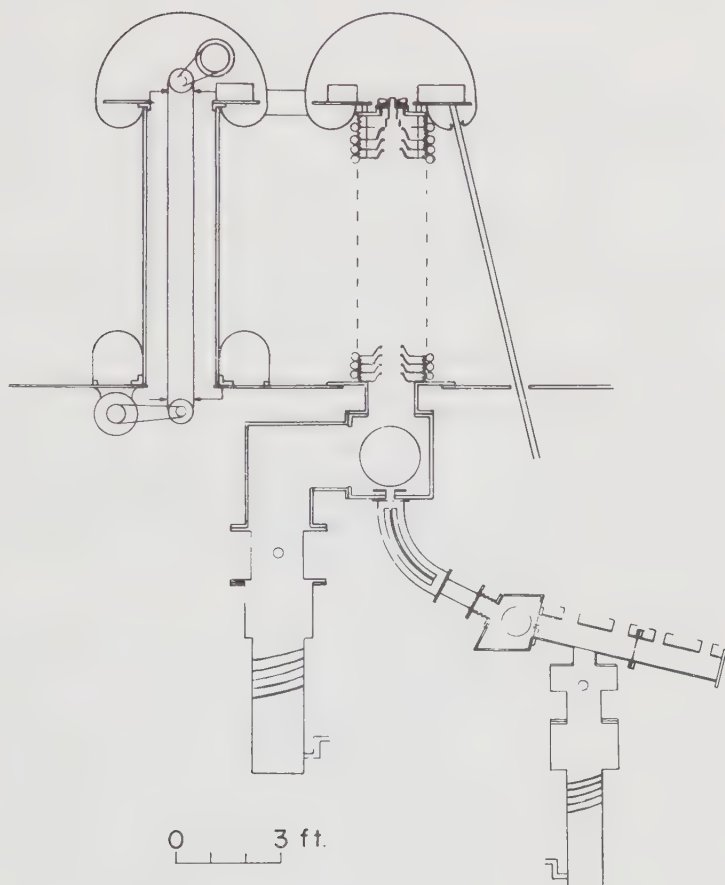


Figure 43. 500 kev Van de Graaff positive ion accelerator with attachments.

## 7. Total Cross Sections for Multiple Ionization in Single Collisions

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University of Connecticut

Total cross sections for the multiple ionization of an ion in single collisions with an atom may evidently be obtained by integrating the measured differential cross sections over all angles of scattering. Previous differential measurements of single collisions of helium, neon, and argon ions and atoms at energies of 25 to 100 keV have been reported by this laboratory in two papers.<sup>78, 79</sup> Similar studies have been reported by Fedorenko<sup>80</sup> and by Kaminker and Fedorenko.<sup>81</sup> These investigations have shown that for the higher charge states important contributions to total cross sections arise from large-angle scattering.

Measured values of each of the total cross sections have been obtained for processes in which a singly ionized atom strikes a neutral target atom, and if the projectile ion is of sufficiently high  $Z$ , it may be 0, 1, 2, 3, ..., 7 times ionized after the collision. These are called respectively the cross sections for electron capture (sometimes called charge exchange), "elastic" scattering, and various degrees of multiple ionization. The collisions studied include  $\text{He}^+$  incident on He, Ne, and A;  $\text{Ne}^+$  on Ne and A; and  $\text{A}^+$  on A at 25, 50, and 100 keV energies. The electron-capture cross sections have been compared with those of a number of other investigators. In the case of "elastic" scattering (scattering without change of charge), the cross section for scattering in excess of  $1^\circ$

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<sup>78</sup>Carbone, Fuls, and Everhart, Phys. Rev. 102, 1524 (1956).

<sup>79</sup>Fuls, Jones, Ziemba, and Everhart, Phys. Rev. 107, 704 (1957).

<sup>80</sup>N. V. Fedorenko, Zhur. Tekh. Fiz. USSR 24, 784 (1954).

<sup>81</sup>D. M. Kaminker and N. V. Fedorenko, Zhur. Tekh. Fiz. USSR 25, 2239 (1955).

was measured. For some of the higher charge states, Kaminker and Fedorenko<sup>81</sup> show data on the cross sections for "stripping" which may be compared with our results.

The procedure is to determine the contribution to the total cross section arising from each of the several angular regions into which the incident particle may be scattered. The differential cross section measurements for the  $1^\circ$  to  $4^\circ$  region were published in our 1957 paper.<sup>79</sup> It was necessary to take angular data closer to the direct beam for the purpose of integration, since several of the differential cross sections rise rapidly near the forward direction. Slight modifications in the apparatus were required for the

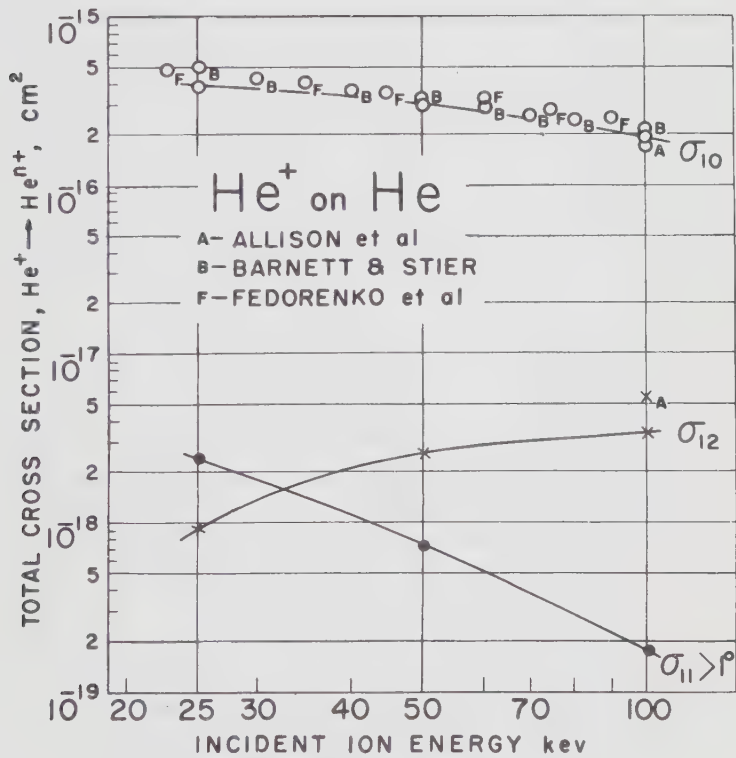


Figure 44. Cross sections for He<sup>+</sup> in the target gas helium obtained by integrating angular distributions and compared with the data of other observers.

measurements from 0 to 1 degree. By integrating the measured values of  $d\sigma/d\Omega$  using the formula

$$\sigma_{if} = 2 \int_0^\pi \left[ d\sigma_{if}/d\Omega \right] \sin\theta d\theta$$

approximately 80 cross sections have been obtained as functions of energy between 20 and 100 kev. The projectile ions and the target gases were of the noble gas group. Integrated cross sections range from  $140 \times 10^{-17} \text{cm}^2$  for  $\sigma_{10}$  for  $A^+$  in A gas at 25 kev, to  $0.0014 \times 10^{-17} \text{cm}^2$  for the creation of 5 times ionized neon in collisions of  $\text{Ne}^+$  in neon gas at 100 kev.

Figure 44 illustrates the extent of the agreement of our integrated cross sections with the work of others, in a case where comparison is possible. It is considered that the agreement is satisfactory. In Figure 44,  $\sigma_{11}$  is the cross section for scattering without change of charge, integrated over all angles greater than  $1^\circ$ .



## IV. ATOMIC AND MOLECULAR SCATTERING

### 1. Experimental Cross Sections of the Free Hydrogen Atom

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Using a modulated atomic beam technique, the cross sections for several types of collisions involving the free hydrogen atom and either electrons or ions have been measured. In these experiments the charged particle beam, which is run dc, crosses the neutral particle beam which is mechanically chopped at a low frequency. With this arrangement any signal developed because of the interaction of the two beams is separable from the much larger signals arising from collisions of the charged particle beam with the residual gas in the vacuum chamber, because the latter is dc (plus noise) while the former occurs at the modulation frequency and in a specific phase.

In the studies of electron collisions with hydrogen atoms, for the ionization cross section, a simple mass spectrometer detected the ions formed in the collisions; for excitation of Lyman alpha radiation, and iodine-vapor-filled ultraviolet photon counter was used; and for elastic scattering, the scattered electrons were detected. In the measurement of charge exchange between both protons and molecular ions and hydrogen atoms, both the mass spectrometer and the customary nondiscriminating slow ion detectors were used.

Since the wave functions of the hydrogen atom are known, comparison of these results with theoretical predictions gives direct information on the validity of the various scattering approximations which have been employed in the theory.

Results of this work have been published subsequently in three articles dealing with the ionization,<sup>82</sup> excitation of Lyman alpha radiation,<sup>83</sup> and elastic scattering<sup>84</sup> in electron-hydrogen atom collisions, and in one article dealing with charge exchange<sup>85</sup> in proton-hydrogen atom collisions.

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<sup>82</sup>Wade L. Fite and R. T. Brackmann, Phys. Rev. 112, 1141 (1958).

<sup>83</sup>Ibid., 1151 (1958).

<sup>84</sup>R. T. Brackmann, Wade L. Fite and Roy H. Neynaber, Phys. Rev. 112, 1157 (1958).

<sup>85</sup>Wade L. Fite, R. T. Brackmann and William R. Snow, Phys. Rev. 112, 1161 (1958).

## 2. Charge Transfer in Atomic and Molecular Hydrogen

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In assessing theoretical predictions of ionization, charge-transfer, and other inelastic cross sections, we must bear in mind (a) that every inelastic collision involves at least three fundamental particles, and (b) that with presently available high-speed computers there is no three-or-more-particle collision of physical interest for which an exact treatment has been practical. Two-particle collisions can be treated exactly, however, using modern computers, even for very complicated interactions. Consequently, experience with two-particle systems, wherein collisions always are elastic, often provides the sole justification for approximations employed in inelastic-collision problems. In actual application, moreover, approximation errors have been estimated either very crudely or not at all, so that reasonable limits of error on the theoretical predictions come only from direct comparison with experiment. For all these reasons, our intuitions concerning inelastic processes are not as firmly grounded as our grasp of two-particle collisions, and we should avoid being over-confident of our understanding of any particular reaction, even when there is apparent agreement between theory and observation.

We shall illustrate the foregoing general remarks by examining closely two of the simplest charge-transfer reactions, namely, protons incident on Atomic H,



and protons incident on molecular  $H_2$  to form  $H_2^+$



The atomic charge-transfer reaction is illustrated in Figure 45. Proton 1 is incident along  $n_i$  and impinges on a stationary hydrogen atom composed of electron  $e$  and proton 2. The electron is captured, forming a hydrogen atom with 1, which goes out along direction  $n_f$ . In the first Born approximation, the differential cross section  $d\sigma$  for the process is

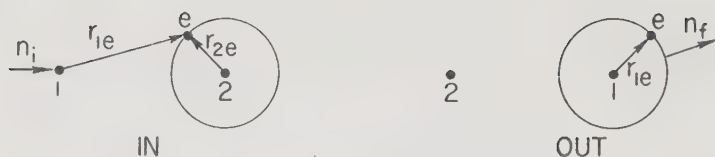


Figure 45. Diagrammatic illustration of charge transfer in atomic hydrogen.

$$d\sigma(\tilde{n}_f) = \left( \frac{\mu}{2\pi\hbar^2} \right)^2 \left| \langle X_f | V_{1e} + V_{12} | X_i \rangle \right|^2 \quad (2a)$$

$$H = T_1 + T_2 + T_e + V_{12} + V_{1e} + V_{2e} \quad (2b)$$

In equation (2a),  $\mu \approx 1/2$  is the reduced mass and  $X_i$  and  $X_f$  are the initial and final wave functions.  $X_i$  is a plane wave times a ground-state hydrogen wave function for electron  $e$  relative to proton 2; similarly,  $X_f$  is a plane wave times a hydrogen wave function of  $r_{1e}$ , the distance between proton 1 and  $e$ . The potentials  $V$  in the Hamiltonian are Coulomb interactions; for instance,  $V_{12} = e^2/r_{12}$ . The cross section is expressed in terms of the matrix element of the "prior" interaction seen by the incident proton, namely,  $V_{1e} + V_{12}$ .

An electron in the first Bohr orbit has a velocity  $c/137 =$  about  $2 \times 10^8$  cm/sec; a proton of this speed has an energy of about 25 kev. At incident proton energies exceeding 25 kev, where the first Born approximation is expected to be valid, there are no data for capture in atomic hydrogen. There are measurements of capture in molecular hydrogen, however, and it has seemed reasonable to assume that at high incident proton energies one  $H_2$  molecule is equivalent to two H atoms for the purposes of charge transfer. The dots in Figure 46 are the experimental points of Barnett and Reynolds<sup>86</sup> for charge transfer in  $H_2$ , expressed per atom of hydrogen; in other words, the dots are half the actually observed charge-transfer cross sections in  $H_2$ . The solid line is the cross

<sup>86</sup>Barnett, C. F., and H. K. Reynolds, "Charge Exchange Cross Sections of Hydrogen Particles in Gases at High Energies," *Phys. Rev.* 109, 355 (1958).

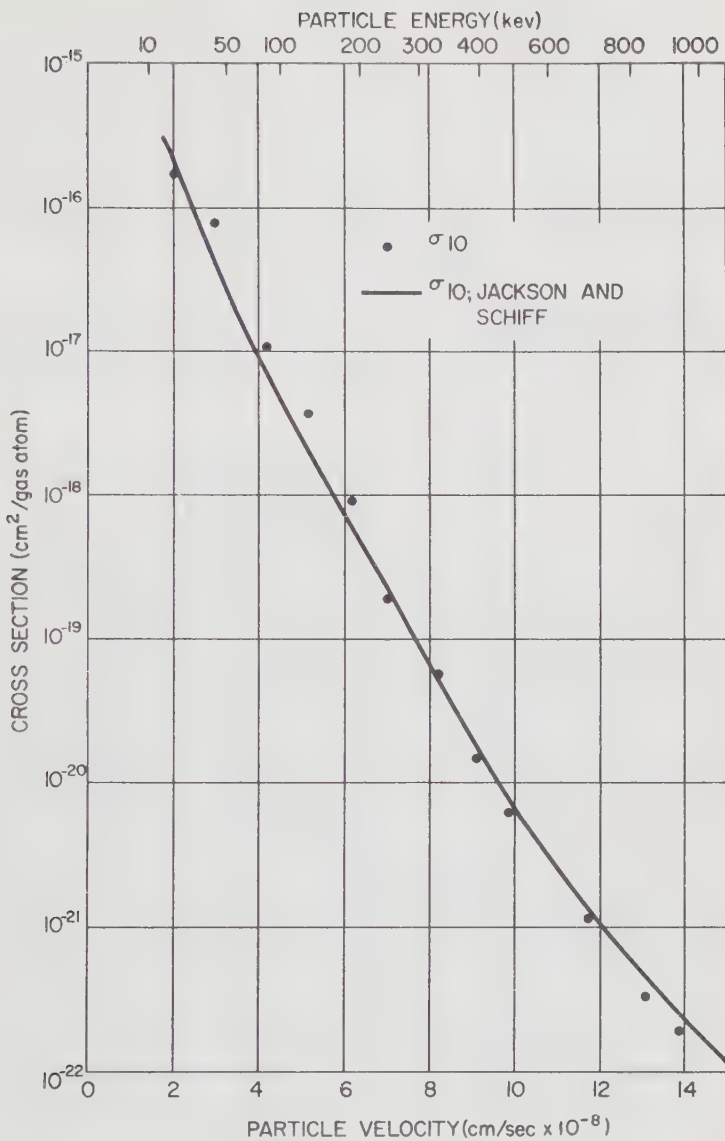


Figure 46: Experimental and theoretical curves for charge transfer in hydrogen.



section predicted in the first Born approximation, as computed by Bates and Dalgarno<sup>87</sup> and, at about the same time, by Jackson and Schiff.<sup>88</sup> Actually, these authors have computed capture probabilities not only into the ground state of hydrogen, as in equation (2a), but also into excited states. It turns out that the excited states make a relatively small contribution to the total charge-transfer cross section, which contribution is almost indiscernible on the log scale of Figure 46. For this reason, we shall simplify our discussion by not distinguishing between the total cross section and the cross section for capture into the ground state only. We note further that the atomic charge-transfer reaction is so symmetric that no "post-prior" discrepancy<sup>89</sup> can occur; in this especially simple case, the expressions for the post and prior matrix elements are completely identical.

This verification of the first Born approximation for charge transfer in atomic H is well known and appears to indicate that we have a satisfactory theory at energies of about 40 kev. However, before relying on this apparent agreement between theory and experiment, we must take a harder look at the assumptions we have made. In the first place, is it legitimate to compare the measured cross section per atom in molecular hydrogen with the predicted cross section for atomic hydrogen? In Figure 47 we illustrate charge transfer in  $H_2$ , with proton 1 coming in and capturing electron A. The initial  $H_2$  molecule is composed of electrons A, B and protons 2, 3. Of course the electrons really are indistinguishable, but it can be shown rigorously that the true cross section for electron capture in this molecular reaction is precisely twice the cross section for capture of electron A, computed as if the electrons were distinguishable. Thus, the differential cross section  $d\sigma$  is

$$d\sigma(n_f) = 2 \left( \frac{\mu}{2\pi\hbar^2} \right)^2 \left| \langle X_f | V_{1A} + V_{12} + V_{13} + V_{1B} | X_i \rangle \right|^2 \quad (3a)$$

<sup>87</sup>D. R. Bates, and A. Dalgarno, "Electron Capture. III. Capture into Excited States in Encounters between Hydrogen Atoms and Fast Protons," Proc. Phys. Soc. (London), 66A, 972 (1953).

<sup>88</sup>J. D. Jackson and H. Schiff, "Electron Capture by Protons Passing through Hydrogen," Phys. Rev. 89, 359 (1953).

<sup>89</sup>H. S. W. Massey, Handbuch der Physik, 36, 282 (1956). Springer-Verlag, Berlin.

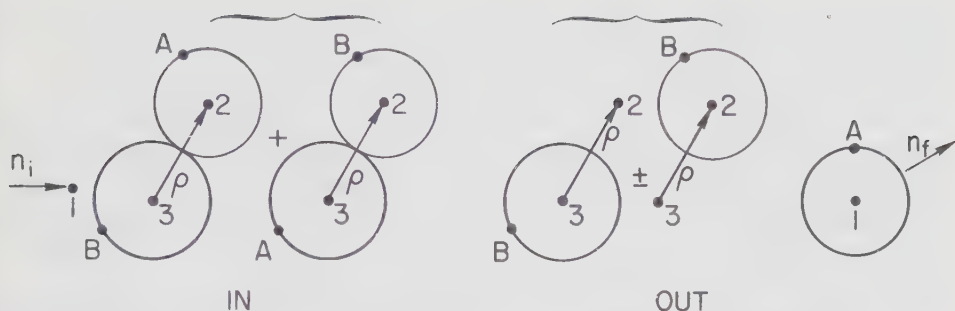


Figure 47. Diagrammatic illustration of charge transfer in molecular hydrogen.

$$\begin{aligned}\phi_i &\sim u(A2)u(B3) + u(A3)u(B2) \\ \phi_f &\sim u(A1) [u(B3) \pm u(B2)]\end{aligned}\quad (3b)$$

As previously, equation (3a) gives  $d\sigma$  in first Born approximation and employs the matrix element, between initial and final states, of the prior interaction seen by proton 1. The functions  $\phi$  are the bound-state parts of  $X_i$  and  $X_f$ . Since the aforementioned factor 2 has been included in equation (3a), the matrix element is for capture of electron A only, as is seen from Figure 47 and from the form for  $\phi_f$ . The  $H_2$  wave function  $\phi_i$  and Figure 47 correspond to the Heitler-London, or extreme atomic orbital, approximation. However, the results we shall quote are essentially the same using either the extreme molecular orbital approximation for  $H_2$  or the intermediate so-called Weinbaum approximation.<sup>90</sup> Recognizing that on the average electron A spends half its time in the vicinity of proton 2 and half its time in the vicinity of proton 3, it is clear that in our comparison of charge transfer in atomic and molecular hydrogen we are making the following two assumptions: first, that the charge cloud around 2 is very much like the electron distribution in a hydrogen atom; and second, that the charge clouds around each nucleus act like independent atoms.

These two assumptions can be stated more precisely as: first, that the amplitude for capture when A is in the vicinity of 2 is the

<sup>90</sup>L. Pauling, and E. B. Wilson, "Introduction to Quantum Mechanics," pp. 340-349, especially p. 346 (1935), McGraw-Hill Book Company, Inc., New York.

same as the amplitude for capture from an isolated hydrogen atom; and second, that the amplitude for capturing A from the cloud around 2 does not interfere with the amplitude for capturing A from the cloud around 3. Put this way, it now can be seen that the comparison of theory for atomic charge transfer with experiments for molecular charge transfer is quite unreasonable. In fact, there are no less than four different effects causing capture from the molecule to differ from capture from isolated H atoms. We shall try to describe these effects without going into too much detail.

First, at high energies the amplitude for capture from an isolated atom<sup>88, 91</sup> is approximately proportional to the Fourier transform of the atomic wave function at the velocity of the incident proton; in other words, the capture amplitude squared is approximately proportional to the probability that the incident proton will find the electron moving along with it, under which circumstance capture is facilitated. As a result, the atomic capture cross section is approximately proportional to  $Z^5$ , where  $Z$  is the nuclear charge on the original atom. With increased  $Z$  the atom is more tightly bound, and the wave function has much larger high-momentum components. The effective nuclear charge seen by an electron in  $H_2$  is<sup>90</sup> about 1.17, rather than unity as in atomic H:  $(1.17)^5 = 1.9$ . Consequently, at very high energies this effect, if uncompensated, would make the capture cross section per atom of  $H_2$  about twice as large as the capture per atom of H. At more moderate energies, where the proton velocity is not much larger than  $2 \times 10^8$  cm/sec, this effect is less important.

To understand the second effect, we must recognize that in equation (2a) for the atomic reaction, the two different terms have different physical interpretations. The matrix element of  $V_{1e}$  represents capture of the electron as a result of the interaction between the electron and the incident proton--we may call this the Brinkman-Kramers term;<sup>91</sup> the matrix element of  $V_{12}$  represents capture of the electron as a result of the interaction between the two protons or (to put it differently) of momentum transfer between two protons. We shall call this the Jackson-Schiff term,<sup>88</sup> but we could equally well have called it the Bates-Dalgarno term.<sup>87</sup> Now we turn to equation (3), and concentrate for the moment on the

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<sup>91</sup>H. C. Brinkman, and H. A. Kramers, "Theory of the Emission of Electrons by  $\alpha$ -particles," Proc. Acad. Sci. Amsterdam, 33, 973 (1930).

$u(A2)$  terms in the matrix element (coming from the factor  $\phi_1$  in  $X_i$ ), remembering that the factor 2 in equation (3a) allows us to compute as if the electrons A, B were in fact distinguishable. We see that for these  $u(A2)$  terms, of the four interactions in the matrix element,  $V_{1A}$  corresponds to the atomic Brinkman-Kramers term.  $V_{12}$  corresponds to capture of A as a result of the interaction between the incident proton and the proton about which A is momentarily orbiting; thus,  $V_{12}$  here is the molecular analogue of the Jackson-Schiff term. On the other hand,  $V_{13} + V_{1B}$  corresponds to capture of A from one atom as a result of the interaction between the oncoming proton and the other atom. Such matrix elements have no analogue in the atomic case. Of course, we must also include the terms involving  $u(A3)$ . For these, we again interpret  $V_{1A}$  as the Brinkman-Kramers term, but now  $V_{13}$  is the Jackson-Schiff term, and  $V_{12} + V_{1B}$  are the nonanalogous interactions corresponding to the "other atom." These nonanalogous terms yield very complicated integrals, difficult to evaluate accurately even in the very high energy limit. It appears, however, that the nonanalogous terms are not negligible, although they are smaller than the terms we have dubbed analogous; also, the nonanalogous terms tend to reduce the molecular amplitude as compared with the atomic case.

These two effects bear on the first assumption discussed above, as they indicate that the amplitude for capture when A is in the vicinity of one of the nuclei, say proton 2, need not be the same as the amplitude for capture from an isolated hydrogen atom. Whether or not these amplitudes are the same, the next two effects show that our second assumption is violated--the charge clouds around 2 and 3 do not act like independent atoms. We mention first an effect which depends on the detailed properties of the molecular wave functions. As shown in Figure 47 and equation (3b), there are two possible final states in  $H_2^+$ --gerade, corresponding to the plus sign, and ungerade, associated with the minus sign. If the internuclear separation  $\rho$  were infinite, the probabilities of capture into either of these states would be equal (as we must expect since at infinite separation the molecule is no different from two isolated H atoms). At the actual internuclear separation ( $0.74 \text{ \AA}$  in  $H_2$ ), however, there is a much-reduced matrix element for transition between the gerade ground state of  $H_2$  and the ungerade state of  $H_2^+$ . This result of the theory is supported by Keene's<sup>92</sup> mass-spectroscopic

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<sup>92</sup>J. P. Keene, "Ionization and Charge Exchange by Fast Ions of Hydrogen and Helium," *Phil. Mag.* 40, 369 (1949).



analysis of the slow ions produced in the reaction (1b). At 15 kev, admittedly a somewhat low energy for this discussion, Keene found very few protons; protons are an inevitable dissociation product of the repulsive ungerade  $H_2^+$  state. The effect we are describing in this paragraph tends to reduce the  $H_2$  capture cross section per atom relative to capture in H, because the capture probability into the gerade state is not much bigger at 0.74 Å than at infinite internuclear separation.

Finally, there is a true interference effect, which can be understood very easily for the so-called analogous terms, wherein the capture amplitude from the cloud around 2 does not involve interactions with the cloud around 3. Suppose we have two scattering centers located at 2 and 3, a distance  $\rho$  apart, with individual scattering amplitudes  $A_2$  and  $A_3$ , as shown in Figure 48. Then classically, if a wave is incident with momentum  $\underline{K}_i$  and goes out with momentum  $\underline{K}_f$ , the extra path covered by the ray scattered by 2 is  $02$  propagating with  $\underline{K}_i$ ;  $3P$  is the extra path covered by the ray scattered by 3, propagating with  $\underline{K}_f$ . Consequently, the amplitudes  $A_2$  and  $A_3$  are related by

$$A_2 = A_3 \exp [i(\underline{K}_i - \underline{K}_f) \cdot \underline{\rho}] \quad (4)$$

Quantum mechanically, taking the origin at 3, the scattering amplitude  $A_3$  is the matrix element of the potential  $V(\underline{r})$  between the c initial and final plane wave states  $\exp(i\underline{K}_i \cdot \underline{r})$  and  $\exp(i\underline{K}_f \cdot \underline{r})$ , i. e.,

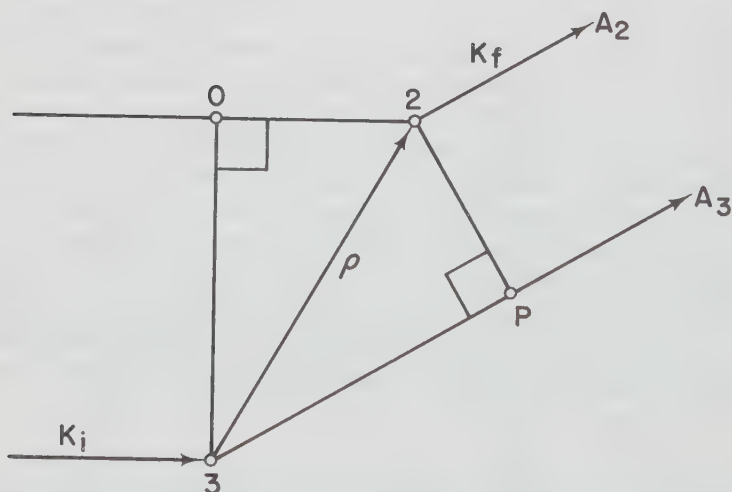


Figure 48. Scattering of a plane wave by two centers.



$$A_3 = \int d\mathbf{r} e^{-i\mathbf{K}_f \cdot \mathbf{r}} V(\mathbf{r}) e^{i\mathbf{K}_i \cdot \mathbf{r}} \quad (5a)$$

The amplitude  $A_2$  is the same except that the potential is  $V(\mathbf{r} - \boldsymbol{\rho})$ , i. e.,

$$A_2 = \int d\mathbf{r} e^{-i\mathbf{K}_f \cdot \mathbf{r}} V(\mathbf{r} - \boldsymbol{\rho}) e^{i\mathbf{K}_i \cdot \mathbf{r}} \quad (5b)$$

Translating the origin in equation (5b) to 2, we again obtain the relation (4). This deduction of the phase relation between the two amplitudes is for potential scattering, but it can be seen by precisely the same kind of argument that the identical relation holds for capture of an electron from an atom centered at 2, as compared with capture from an atom centered at 3.

Naive considerations might lead us to think that the phase relation we have demonstrated is unimportant in the energy range above 25 kev, where the wave length of the incident proton is so much smaller than the internuclear spacing (0.74 Å). In other words, because  $K_i$  is a large number, we might think the interface would wash out. The effective phase factor is not  $K_i \rho$ , however, but  $(K_i - K_f) \cdot \boldsymbol{\rho}$ , i. e.,  $\rho$  times the momentum change, and this is a very much smaller number than  $K_i \rho$  because in picking up the very light electron the incident proton hardly is deflected. This classical argument is borne out by examination of the quantum mechanical matrix elements, and in fact there is effective capture only into angles such that  $|K_i - K_f| \sim (m/M) K_i$ . Consequently, the effective wave length for the interference in this charge-transfer process is not the wave length of a proton at the incident proton velocity, but the 2000-times-larger wave length of an electron at that velocity. Of course, it is necessary to average over-all orientations of  $\boldsymbol{\rho}$ , but this can be done, and it turns out that for capture by the bound gerade  $H_2^+$  state the interference is significantly constructive at energies less than 400 kev. Above 400 kev the interference first becomes destructive, then rapidly washes out. Thus, this effect is important mainly at moderate energies, where it tends to increase the molecular cross section relative to the atomic cross section.

From the discussion so far, we are entitled to draw the following conclusion about the experimental points plotted in Figure 46. There are several compensating effects which are difficult to evaluate precisely, but if the molecular capture cross section per atom does equal the atomic capture cross section, this equality must be fortuitous. Moreover, whether or not we believe these experimental points have anything to do with charge transfer in atomic

hydrogen, there are strong reasons to disbelieve the theoretical curve. To make our position completely clear: The theoretical curve of Figure 46 was computed in the first Born approximation, and we feel that for charge-transfer collisions, which are so different in character from potential scattering, the first Born approximation need not be valid at high energies. Before explaining the reasons for this assertion, we remark that in questioning the first Born approximation we are questioning the quantitative details of the effects in  $H_2$  which were discussed above, since this discussion was based on the first Born approximation for the molecular matrix elements. However, some of the effects were so understandable physically--the interference especially--that they should be predicted by an exact calculation.

For the purpose of discussing the validity of the first Born approximation in charge transfer, it is sufficient to consider the simplest case, namely, capture by protons in atomic hydrogen. We observe that in the first Born approximation expression (2a) for the cross section, the matrix elements of  $V_{1e}$  and of the proton-proton interaction  $V_{12}$  costar with equal billing. This seems surprising, since one would guess that the capture cross section would be pretty much the same even if the protons did not interact. Seemingly, the proton-proton interaction serves merely to slightly deflect the fast incident proton, and this slight deflection hardly should effect the probability of electron capture. This was the point of view of Brinkman and Kramers,<sup>91</sup> who gave the first acceptable quantum mechanical treatment of this charge-transfer reaction. They considered the proton-proton interaction inconsequential, dropped it from the Hamiltonian, and then obtained the cross section by first-order time-dependent perturbation theory. This procedure neglects distortion of the true wave function from its initial or final plane wave forms, which is precisely the assumption made in the first Born approximation, so it is not surprising that Brinkman and Kramers deduce precisely the cross section one would get in the usual first Born approximation if the  $V_{12}$  term had been left out of the Hamiltonian from the beginning. In other words, Brinkman and Kramers got the result obtained by putting  $V_{12} = 0$  into equation (2a). In addition to the derivation just described, Brinkman and Kramers gave another deduction of their cross section. They argued that since the protons were so heavy as compared with the electrons, one could simply replace the protons by classical centers of force, reducing the original three-body problem to the problem of a single quantum mechanical particle, the electron, under the influence of a time-dependent interaction  $e^2/r_{1e}$ . The time-dependence of  $r_{1e}$  is known at each impact parameter, assuming

$r_1$  moves in a straight line with speed  $v$ . Computing the cross section at each impact parameter by first-order time-dependent theory, as before, and summing over-all impact parameters, one gets the previous result of Brinkman and Kramers. As Drisko<sup>93</sup> has pointed out, this second derivation has the important feature that it is not necessary to neglect the proton-proton interaction: With the protons no longer quantum mechanical particles but merely centers of force, it can be proved rigorously that the electron capture cross section is independent of the proton-proton interaction.

The point of this discussion is that in the first Born approximation the  $V_{12}$ , or Jackson-Schiff, matrix element very nearly equals the  $V_{1e}$ , or Brinkman-Kramers, matrix element, despite the above appealing arguments for thinking that the  $V_{12}$  contribution should be small. This certainly suggests strongly that the first Born approximation is seriously wrong. There are other arguments against the correctness of the first Born approximation which we shall list briefly. First, the impact-parameter treatment<sup>91, 93</sup> shows that with increasing proton energy the cross section results from capture at smaller and smaller impact parameters; in fact, at infinite energy only protons of zero impact parameter produce significant capture. This is contrary to physical expectations, as there seems no reason why capture should not occur for any proton passing through the electron cloud. In other words, the significant impact parameters should be energy independent and should not be vanishingly small compared with a Bohr radius. Second, it can be argued that fast incident protons are essentially classical, which is the content of the Brinkman-Kramers approach, and therefore that a correct high-energy approximation should agree with a classical calculation. It happens that there is a classical calculation of the cross section by L. H. Thomas<sup>94</sup> who obtained a  $v^{-11}$  dependence of the cross section at high energies. Both the Jackson-Schiff and Brinkman-Kramers first Born calculations predict a  $v^{-12}$  dependence.

This last result is interesting, since it suggests that to get a correct answer it is not enough simply to drop the proton-proton interaction in the first Born approximation. The most obvious next step is to try the second Born approximation. Unfortunately, there is the customary difficulty that the second Born approximation

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<sup>93</sup>R. Drisko, Ph. D. thesis (unpublished), Carnegie Institute of Technology (1955).

<sup>94</sup>L. H. Thomas, Proc. Roy. Soc. (London), A 104, 561 (1927)



matrix elements are very hard to evaluate, even in the high-energy limit; in fact, there is some disagreement concerning the value of this limit. Jackson<sup>95</sup> claims the second Born approximation is very small compared with the first Born terms; Drisko<sup>93</sup> claims the second Born terms dominate the first Born terms at very high energies. Both Drisko and Jackson must make approximations to obtain their results; moreover, the expressions from which they start, though equivalent if one could make an exact calculation, are not necessarily identical in an approximate calculation. It is this writer's opinion that Drisko is correct, because his results do agree with physical expectation. First, his cross section varies as  $v^{-11}$ , in agreement with Thomas's classical result. Second, the impact parameters which contribute in his second Born approximation are of the order of the Bohr radius. Third, in his second Born approximation the proton-proton interaction term is unimportant, as we argued it should be.

Jackson is not altogether wrong, however--the energy at which the second Born term starts to dominate the first is very high, being hundreds of Mev, according to Drisko. Thus, after this long discussion we find ourselves concluding, rather unexpectedly, that the first Born approximation dominates and may well be correct at intermediate energies, although it fails in the high-energy limit where it usually is supposed to hold. This conclusion leaves the question: How can the first Born approximation be correct when its matrix elements and impact-parameter dependence are so different from what one expects in a classical description? As yet there is no satisfactory answer to this question, and until it is answered we cannot claim to understand charge transfer at intermediate energies. The writer will hazard that the classical description is incorrect at intermediate energies for two reasons: First, as explained previously, the momentum change during electron capture is so small that the effective wave length is 2000 times as large as one would suppose, which means that the classical limit--the wave length small compared with size--is not attained until the incident proton energy is much higher than one would suppose. Second, in Coulomb interactions the significant parameter,<sup>96</sup> in elastic scat-

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<sup>95</sup>J. D. Jackson, "On the Use of the Complete Interaction Hamiltonian in Atomic Rearrangement Collisions," Proc. Phys. Soc. (London), A 70, 26 (1957).

<sup>96</sup>D. Bohm, Quantum Theory, p. 554 (1951), Prentice-Hall, Inc., New York.

scattering at any rate, is  $e^2/\hbar v$ , and the classical limit  $\hbar \rightarrow 0$  corresponds to  $v \rightarrow 0$ , not to  $v \rightarrow \infty$ . By invoking this argument, based on elastic scattering, we are doing exactly what we warned against at the beginning of this paper, but it may be that this feature of Coulomb interactions helps to extend the energy region in which the classical description is not correct.

We shall conclude by listing a few experiments which are suggested by this discussion of charge transfer in atomic and molecular hydrogen. Clearly, it is extremely worthwhile to extend the atomic charge-transfer measurements<sup>97</sup> to higher energies, well beyond the Dalgarno-Yadav<sup>98</sup> perturbed stationary-state region. These measurements would first give a direct and unequivocal test of the first Born approximation theories, both of Jackson and Shiff and of Brinkman and Kramers. If these experiments are accurate enough and are carried to high energies, it may be possible to see if the energy dependence really is  $v^{-12}$ , as is predicted by both of these theories. On Drisko's theory, the  $v^{-11}$  classical dependence occurs at energies too high to be interesting, and much higher than those that have been reached. In this connection it is of interest, therefore, that Barnett and Reynolds' experimental points (Figure 46) have a  $v^{-10}$  dependence. It is of further interest that many of the experiments, for instance those of Barnett and Reynolds, do not distinguish between a true charge transfer and an elastic collision with large momentum transfer, producing a slow ion and a fast neutral. The cross section for such collisions is unimportant at low energies but decreases much less rapidly than  $v^{-11}$  or  $v^{-12}$ , so that such collisions necessarily dominate charge transfer at sufficiently high energies and must be guarded against. More details cannot be given here because the energy at which elastic collisions become important depends on the minimum elastic momentum transfer which will be interpreted as electron capture in the particular experimental setup. Using incident deuterons on atomic hydrogen, and mass analysing the slow ions formed, enables a direct measurement of this large-momentum-transfer elastic cross section and of the true charge transfer. Furthermore, comparison of the charge

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<sup>97</sup>W. L. Fite, R. T. Brackmann, and W. R. Snow, "Charge Exchange in Proton-Hydrogen-Atom Collisions," General Atomic Report GA-407, June 26, 1958 and Ref. 85.

<sup>98</sup>A. Dalgarno, and H. N. Yadav, "Electron Capture. II. Resonance Capture from Hydrogen Atoms by Slow Protons," Proc. Phys. Soc. (London), A 66, 173 (1953).



transfer cross section with incident deuterons and protons on atomic hydrogen could yield some interesting information on proton exchange during the charge-transfer process. (It will be noted that proton exchange was entirely neglected in this discussion.) Extension of the atomic-hydrogen charge-transfer measurements to higher energies also will check the hypothesis that for purposes of charge transfer one  $H_2$  molecule is equivalent to two H atoms. Figure 46 suggests that this hypothesis may hold for hydrogen, but we have claimed that if so it must be fortuitous. Comparison of other atoms and molecules, e. g., of atomic and molecular oxygen, should shed further light on the generality of the hypothesis. Finally, careful charge-transfer experiments on molecules ought to manifest the interference effect, which seems to be understandable on very general grounds and which apparently depends only on the incident energy, the internuclear distance, and whether or not the final molecular ion is in a gerade or ungerade state (which determines whether the interference is constructive or destructive). These features of the interference effect should hold for most homonuclear diatomic gases, not just hydrogen. However, distinguishing experimentally between transitions to gerade and ungerade states may not be as easy with other molecules as with hydrogen, where  $H_2^+$  ions result from transitions to gerade states only, while slow protons are produced in large degree (though not solely) by transitions to ungerade states.

We wish to acknowledge numerous conversations with Dr. Richard Drisko. We also thank Robert Bassel and Tai-Fu Tuan, on whose thesis, in preparation for the University of Pittsburgh, much of this discussion is based.

## V. IONIZATION IN GASES BY HIGH-ENERGY PARTICLES

### 1. Total Ionization in Gases by High-Energy Particles: An Appraisal of our Understanding

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The efficiency of ionization of gases by swiftly moving charged particles is generally measured by  $W$ , the mean energy expended per ion pair produced. (Quantities analogous to  $W$  can be determined in many solids,<sup>99</sup> but they will not be considered here.) For complete absorption, the most common case,  $W = T_0/N_i$ , where  $T_0$  is the initial kinetic energy of the particle and  $N_i$  is the average total number of ion pairs formed. (More accurately,  $\pm N_i e$  is the total electric charge of either sign; this allows for the formation of multiply charged positive ions.) Measurements of  $W$  have been made with abundance since the earliest days of radioactivity. The pre-eminent pioneer in this field was Sir W. H. Bragg, whose 1912 book is still a valuable source of information and insight. The voluminous literature that has accumulated since then attests to the unfortunate fact that experimental determination of  $W$  is often very easy, but accurate determination is not. This literature abounds in disparities and contradictions, and even the greatly augmented activity of the past decade has failed to resolve many of them.

Experimental values of  $W$  for alpha or for beta particles in most gases lie in the range of 20 to 40 ev/ion pair, and may vary with the nature and the energy of the particle. This dependence is in many cases slight, but in some it is marked, and in still others it is apparently absent. The magnitudes of  $W$  are important in a variety of ways. They are particularly vital for many types of nuclear-radiation detectors; with the evolution of these devices the pertinence of  $W$  to certain of them has risen or fallen, but the total interest in

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<sup>99</sup>R. L. Platzman, Chapter 2 in "Radiation Biology & Medicine" (W. D. Claus, ed.); cf. section 2.16. Addison-Wesley Publishing Co., Reading, Massachusetts (1958).

W has never declined. And W-values have much theoretical significance, in part because they are the simplest and most accessible measure of an aggregate effect of high-energy radiation. There is a great deal of physics involved in the absorption of an alpha particle in a gas and usually a great deal of chemistry as well.

The problems that arise in the theoretical interpretation of W may be classified into three distinct categories. The first comprises the individual inelastic collision cross sections. Of chief interest are the total excitation, and the total and differential ionization, cross sections for charged particles (electrons, and such positively charged particles as may be present). In the case of positive ions, the electron-capture and electron-loss cross sections are also involved; the latter very definitely includes the differential loss cross section (i. e., ejected-electron distribution). Angular distributions are not ordinarily relevant, nor are elastic-scattering cross sections.

The second category consists of "bookkeeping problems." The absorption of a single alpha particle, for example, involves some half-million primary elementary processes encompassing many varieties. Thus  $1/W$  is, in effect, an average of the ionization cross section over the entire "degradation spectrum," that is, the spectrum of positively-charged particles and electrons actually present in the medium. (Spencer and Fano, who pioneered in treating these spectra for electron sources,<sup>100</sup> called them "slowing-down spectra.") The bookkeeping problems have not yet been fully explored.

Problems in the third category are in the nature of distractions. Such distractions are of two kinds: trivial ones, which cause errors in the experimental determination of W (and are often not elucidated) and nontrivial ones, which are due to interesting and important physical processes that may alter the apparent value of W. Only the latter, of course, are germane here. Some of them increase, and others decrease the observed value of W.

An increase in W is usually the result of partial recombination of positive ions with negative ions (and perhaps electrons). It is generally believed that recombination is a hazard only in so-called "electronegative" gases (e. g.,  $O_2$ , NO,  $H_2O$ ) and then particularly

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<sup>100</sup>L. V. Spencer and U. Fano, Phys. Rev. 93, 1172 (1954).

for densely ionizing particles, like alpha particles. Such columnar recombination is only imperfectly understood. With nonelectro-negative gases (noble gases,  $N_2$ , hydrocarbons) an ordinary saturation curve is attainable and is believed to ensure against ion loss, although criteria for saturation are by no means unequivocal.

Artifacts that cause a decrease in the apparent value of  $W$  are of at least three types:

a. Ionization of Impurities by Excited Atoms (or Molecules?).

This is the well-known Jesse effect. It has been identified and studied quantitatively only in He and Ne (Jesse) and A (Hurst). For these gases the pertinent excited atoms are long-lived (metastable) and the effect takes over at impurity concentrations greater than about 0.1 mole per cent. In the case of argon the phenomena are somewhat complicated because of the following circumstances:<sup>99</sup> Since the metastable states of A have comparatively low energy, only contaminants with low ionization potential are effective, and these are most conveniently polyatomic molecules--but transfer of energy not vastly in excess of the ionization potential to a polyatomic molecule does not invariably lead to ionization; excitation, followed by dissociation, can compete. (It is well known that a molecule may sustain energy in excess of its ionization potential without losing an electron, the excitation energy ultimately causing dissociation or even, in a large molecule, being partially converted into heat. Direct information on this effect is provided by measurement of the ionization current resulting from absorption of ultraviolet light.<sup>100a</sup> There is, as yet, no quantitative theoretical interpretation of this phenomenon in terms of properties of the excited states involved, nor has it been possible to correlate the results of the optical measurements with the results of Hurst on energy transfer from metastable states of argon. It would be expected that argon should show a normal Jesse effect if monatomic vapors were used as contaminants, but such experiments have not been performed. The existence of the Jesse effect in molecular gases is a remote but most interesting possibility. The effect in cases of short-lived (non-metastable) excited states has not been identified with certainty; the "impurity" concentration here would have to be a few per cent, at the least, and the complications experienced with gaseous mixtures which stem from distortion of the degradation spectrum, would then ensue and becloud the interpretation.

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<sup>100a</sup>G. L. Weissler, Handbuch der Physik, Springer, Berlin, 21, 318 (1956).



b. Ionization of Impurities by Subexcitation Electrons. That portion of the degradation electron-spectrum having kinetic energy less than the lowest excitation potential of the parent gas will have a comparatively protracted existence, and such electrons may therefore ionize impurities with sufficiently low ionization potential.<sup>101</sup> Like many other aspects of inelastic electron collisions in gases, this phenomenon is poorly understood in the case of molecular gases, owing in part to the complications arising from low-lying electronic states of molecules.

c. Ion Production by Formation of Dimeric Molecular Ions. An example is the reaction  $\text{He}^* + \text{He} \rightarrow \text{He}_2^+ + \bar{\epsilon}$ , which occurs with large cross section for all excited states of helium having principal quantum number  $\leq 4$  (and possibly also for  $n = 3$ ). Since this process must compete with allowed radiation of the excitation energy, it suggests<sup>101a</sup> an increase in the apparent  $W$  for pure He by some 7 per cent at low pressures (probably in the range of 1 to 10 cm Hg.); this has not yet been investigated. (The radiation mentioned is from transitions to lower excited states. Transitions to the ground state are, in effect, forbidden because of imprisonment.) The process is well-known in monatomic gases (noble gases, alkali-metal vapors) but unexplored in molecular gases. It might be important for small molecules, but is unlikely to be so for large ones because of the rapid internal dissipation of electronic excitation energy.

There are other latent sources of excess ionization. Among them may be mentioned the collision-induced dissociation of an excited molecule into ionic fragments rather than, in the absence of a collision, uncharged fragments. This possibility, like the processes discussed previously, points to the importance of measuring  $W$  over as great a pressure range as possible--a task that has scarcely been begun.

Let us momentarily pass over the variation of  $W$  with type and energy of particle, and consider the numerical values for the total absorption of alpha and beta particles which are never very different and are a useful standard for discussing those instances in which the  $W$ -value does deviate substantially. For each gas we consider also the ratio  $W/I$  of  $W$  to the ionization potential.

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<sup>101</sup> R. L. Platzman, Rad. Res. 2, 1 (1955).

<sup>101a</sup> R. L. Platzman, Rad. Res. 3, 340 (1955).



Consider first the noble gases. These are the only monatomic gases for which  $W$  has been determined (measurements on metal vapors would be of great interest). For all five noble gases  $W/I = 1.7$  with at most a few per cent divergence. This value may be understood in terms of the energy balance of the absorbed energy  $T_O$ :

$$T_O = N_i \bar{E}_i + N_{ex} \bar{E}_{ex} + N_i \bar{\epsilon}$$

$$W = T_O / N_i = \bar{E}_i + \bar{E}_{ex} (N_{ex} / N_i) + \bar{\epsilon} \quad (1)$$

and

$$W/I = (\bar{E}_i / I) + (\bar{E}_{ex} / I) (N_{ex} / N_i) + (\bar{\epsilon} / I)$$

$$1.70 = 1.06 + (0.85) (0.40) + 0.30$$

There are produced  $N_i$  (ultimately) singly charged atomic ions, at an average energy expenditure of  $\bar{E}_i$ ;  $N_{ex}$  excited atoms at an average expenditure of  $\bar{E}_{ex}$ ; and  $N_i$  subexcitation electrons having average kinetic energy of  $\bar{\epsilon}$ . All the quantities appearing on the right-hand side of this equation can be evaluated from information that is independent of the absolute measurement of  $W$ ,<sup>101a</sup> and completeness of the energy balance can therefore be verified. The quantity 1.06 exceeds unity because of the energy "wasted" in producing excited ions and multiply-charged ions. The quantity 0.85 is an average excitation energy, relative to  $I$ ; it can be estimated successfully because, for the noble gases, all levels lie fairly close to the ionization limit. The quantity 0.40 is the ratio of total number of excited to total number of (singly) ionized atoms produced, and is clearly an average of the ratio of corresponding cross sections over the degradation spectrum of particle energies. It is small because of the well-known characteristic of noble gases that an unusually great proportion of the total oscillator strength resides in the continuum. (The ratio for He of total matrix-element squared to discrete and to continuum states is 0.55.) The quantity 0.30  $I$  is the mean energy, per ion pair, left in the subexcitation electrons and therefore, in the pure gases, dissipated directly as heat. About 18 per cent of the absorbed energy takes this form.

Consider now the molecular gases. For all such gases, including hydrogen,  $W/I$  is much greater than for the noble gases; it lies between 2.1 and 2.6 in most cases. Table XI shows a few selected data. We are not able to interpret these values with anything like the detail possible for the noble gases. The greater magnitude of  $W/I$  has contributions from all of three different factors:

TABLE XI

Selected Values of  $W$ , and of  $W/I$ , for Gases\*  
(Uncertainties are approximately 1 to 2 per cent)

| Gas                           | $\bar{W}_\beta$<br>(beta rays) | $\bar{W}_\beta/I$  |
|-------------------------------|--------------------------------|--------------------|
| He                            | 42.3                           | 1.72               |
| Ne                            | 36.6                           | 1.70               |
| A                             | 26.4                           | 1.68               |
| Kr                            | 24.1                           | 1.72               |
| Xe                            | 21.9                           | 1.80               |
| H <sub>2</sub>                | 36.3                           | 2.35               |
| N <sub>2</sub>                | 34.9                           | 2.24               |
| O <sub>2</sub>                | 30.8                           | 2.55               |
| NO                            | -                              | 3.3(?) ( $W_a/I$ ) |
| air                           | 33.9                           | -                  |
| CO <sub>2</sub>               | 32.8                           | 2.38               |
| CH <sub>4</sub>               | 27.3                           | 2.10               |
| C <sub>2</sub> H <sub>6</sub> | 24.5                           | 2.10               |
| C <sub>2</sub> H <sub>4</sub> | 26.1                           | 2.48               |
| C <sub>2</sub> H <sub>2</sub> | 25.7                           | 2.25               |

\*I is the ionization potential. This table is adapted from reference 99, page 50.

a. Energy Transferred to Molecular Potential Energy. There are two categories here. One, pertinent to stable excited and ionized states, is the extra energy transferred by virtue of the demands of the Franck-Condon principle. For example, the extra energy is small for N<sub>2</sub>, but greater for O<sub>2</sub>, for which (in contrast to N<sub>2</sub>) the upper states have equilibrium nuclear separations markedly different

from that of the ground state. The second, which is much more important, stems from transitions to "repulsive" potential curves (surfaces) for excited and for ionized states. Since the ionization potential refers, by definition, to the lowest electronic state of the molecular ion, inclusion in the averaging of transitions to repulsive states increases the values of both  $\bar{E}_{\text{ex}}/I$  and  $\bar{E}_i/I$ .

b. "Multiple" Electron Transitions. With the noble gases, most of the transitions involve promotion of a single electron; occasionally, however, two or more are excited (as in the case of multiple ionization). Little is known quantitatively about such transitions, especially for molecules; this is particularly so for excitation by electrons having kinetic energy below about several hundred ev, for which excitation cross sections may be comparable to geometrical cross sections. However, there is good reason to believe them to be more common in the case of molecules. Indeed, results from chemical mass-spectrometry have been interpreted as indicating ion formation predominantly in electronically excited states at the bombarding energies commonly used (ca. 70 ev, which is an important although by no means "typical" portion of the degradation spectrum). In the case of certain hydrocarbons, for example, the average electronic excitation energy of the ions has been estimated to lie in the neighborhood of 5 ev.<sup>101b</sup>

This effect obviously increases both  $\bar{E}_{\text{ex}}/I$  and  $\bar{E}_i/I$ .

c. Relative Importance of the Continuum. It is well known that other substances have a smaller proportion of their total oscillator strength in the continuum than the noble gases have. This would lead to values of  $N_{\text{ex}}/N_i$  greater than the one (0.40) found for the latter. One may attempt to compute this ratio from experimental values of  $W/I$ , on the basis of estimated contributions from the other quantities in equation (1). It is found that  $N_{\text{ex}}/N_i$  is about unity for most molecules, but the calculation is very rough. Attention should be directed to interesting regularities in the  $W/I$ -values. For example,  $\text{CH}_4$ , which in some respects is akin to a noble gas atom, has the small value of 2.10, and for other small alkanes it is about the same; adding a double bond in these hydrocarbons markedly increases the value of  $W/I$ . The "hard" molecule  $\text{N}_2$  has an intermediate value, 2.24; the "softer"  $\text{O}_2$  has a much greater one, 2.55. One does not know how to apportion these variations between

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<sup>101b</sup>M. Krauss, A. L. Wahrhaftig, and H. Eyring, Ann. Rev. Nuclear Sci. 5, 241 (1955).

changes in  $\bar{E}/I$ , on the one hand, and changes in  $N_{\text{ex}}/N_i$ , on the other, but the latter is probably the major factor. It is evident that interesting and hitherto unknown optical properties can be revealed by these studies. The  $W/I$  value quoted for NO, an odd-electron molecule which is, in fact, a stable free radical, is very high (3.3); this should be restudied, for NO exhibits very strong electron attachment. Another odd-electron molecule, NO<sub>2</sub>, also has a high value (2.7). Apparently these molecules are similar to atomic hydrogen, for which the ratio of total matrix-element-squared to discrete and to continuum states is 2.53 (compared to 0.55 for helium, mentioned above). In Table XII I have estimated values of  $N_{\text{ex}}/N_i$  for three central cases: atomic helium, molecular hydrogen, and atomic hydrogen. One might surmise that these substances are typical, respectively, of the noble gases, the normal molecules, and the free radicals (molecules containing an odd number of electrons). (Alkali-metal atoms are exceptional: they have an even smaller proportion of their oscillator strength in the continuum.) In addition values of the total matrix-element-squared to discrete states and to continuum states,  $M_{\text{ex}}^2$  and  $M_i^2$ , respectively, are given. In the case of helium,  $M_{\text{ex}}^2$  and  $M_i^2$  are known accurately.<sup>102, 102a</sup>

TABLE XII  
Importance of the Continuum

|                         | He   | H <sub>2</sub> | H      |
|-------------------------|------|----------------|--------|
| $M_i^2$                 | 0.49 | 0.71           | 0.283  |
| $M_{\text{ex}}^2$       | 0.27 | 0.94           | 0.717  |
| $M_{\text{ex}}^2/M_i^2$ | 0.55 | 1.32           | 2.53   |
| $N_{\text{ex}}/N_i$     | 0.50 | 1.1            | (1.8)  |
| $W/I$                   | 1.72 | 2.35           | (2.65) |

<sup>102</sup> W. F. Miller, Ph.D. Dissertation, Purdue University, (1956).

<sup>102a</sup> W. F. Miller and R. L. Platzman, Bull. Am. Phys. Soc. 2, 227 (1957).



The value of  $N_{\text{ex}}/N_i$  inferred from experiment is 0.50, greater than the number 0.40 mentioned earlier because correction has been made for the formation of  $\text{He}_2^+$ . This value is slightly less than  $M_{\text{ex}}^2/M_i^2 = 0.55$  because the probability of excitation is slightly smaller than the asymptotic value (1/1.55) for electrons of all kinetic energies greater than about 100 ev. In the case of  $\text{H}_2$  the values of  $M_{\text{ex}}^2$  and  $M_i^2$  are not known as accurately, and I have calculated them from experimental data as suggested by Fano.<sup>103</sup> Here again the value of  $N_{\text{ex}}/N_i$  inferred from  $W/I$  is slightly smaller than  $M_{\text{ex}}^2/M_i^2$ . For atomic hydrogen, in contrast, the matrix elements are known exactly from theory, but  $W$  cannot be measured; I have used a theoretical calculation of  $W$  which is admittedly not very trustworthy. Altogether, however, the data in the table appear to be consistent and are probably a useful first approach. They suggest that  $N_{\text{ex}}/N_i$  is, respectively, approximately 1/2, 1, and 2, for the three classes of molecules.

We turn now to the variation of  $W$  with energy and type of particle. Experiments with beta rays in several gases, both atomic and molecular, have failed clearly to disclose a measurable dependence of  $W$  upon beta-ray energy; the experiments span the range from 5 to 60-kev mean beta-ray energy, and the lack of energy dependence is established to about  $\pm 2$  per cent. Measurements on the specific ionization at relativistic energies are likewise insufficiently accurate to establish the magnitude of the energy dependence. This is understandable because so much of the measured ionization is caused by the low-energy portion of the degradation electron spectrum, which is comparatively insensitive to the initial electron energy. It would nevertheless be valuable to have measurements sufficiently accurate to reveal and assess the energy dependence, in both atomic and molecular gases. This energy dependence must exist, for the relative probability of excitation and ionization in a single electron collision continues to vary up to the very greatest energies; but since the effect is so strongly mediated by the degradation spectrum, very great experimental accuracy will be required. Experiments with monoenergetic electrons would be ideal, but they are not imminent;  $W$ -measurements with electrons are very difficult, as is evident from Dr. Jesse's description of his painstaking experiments.

The theory of  $W$  for electrons in helium has been developed in detail by Dr. W. F. Miller.<sup>102, 103a</sup> It predicts an energy

<sup>103</sup> U. Fano, Phys. Rev. 95, 1198 (1954).

<sup>103a</sup> W. F. Miller, Bull. Am. Phys. Soc. 1, 202 (1956).



dependence for monoenergetic electrons below about 2 or 3 kev, the value of  $W$  declining with increasing energy and reaching approximate constancy above that region. (This result appears as Figure 2-15 in reference 99.) How molecular gases may behave in this respect is quite unknown.

In the case of alpha particles it is important to distinguish between phenomena occurring near the end of the track, where the particle carries one or two orbital electrons, and those occurring at energies greater than several Mev, where it is certainly a bare nucleus. The most striking experimental fact, not yet elucidated, is that  $W_\alpha/W_\beta$  ( $W_\alpha$  here refers to the total ionization by natural alpha particles) is about 1.05 to 1.07 for all molecular gases except hydrogen, and close to unity ( $\pm 1$  or 2 per cent) for the noble gases and hydrogen (cf. the paper by Jesse which follows). Most, but perhaps not all, of this striking difference stems from the end of the track. It is well known that  $W$  for very slow ions (e. g., alpha-particle recoil atoms) is very much greater than  $W_\alpha$  or  $W_\beta$  because of the large proportion of the particle energy that is transferred to atoms in toto in non-ionizing, quasi-adiabatic "elastic" collisions. However, this non-ionizing fraction (often represented by an "ionization defect," a term I dislike because it assumes more than is in fact known) should, according to simple theory, depend only on the atomic numbers of particle and gas; thus no light is thrown on the cause of the two categories of behavior. Two alternative, possible explanations, both conjectural, are:

a. The  $W$ -value in the capture-loss region,  $W_{cl}$ , is substantially smaller than  $W_\beta$  for the noble gases and hydrogen (this would be the case if loss electrons had very little kinetic energy and there were little likelihood of excitation by neutral particles: then  $W_{cl} \approx I$ ), but  $W_{cl} \approx W_\beta$  for the other molecular gases (extra energy expended in loss collisions and in capture collisions as molecular potential energy, by virtue of interpenetration of "particle" and molecular electron shells). Thus, in the case of the noble gases and hydrogen, the energy "wasted" in nuclear elastic collisions would be compensated, at least in part, by more efficient ion production in the capture-loss region. Hydrogen might fall with the noble gases because it is so small.

b. The charged component of the alpha-particle beam persists to much lower energies in the noble gases and hydrogen than it does in the others. Such would be the case if the cross section for electron capture were smaller at very low energy (say, near 50 kev) in the first gases than in the second. Accordingly, the total energy

"wasted" in nuclear collisions would be smaller for the first group. A possible origin of this distinction is the contribution of molecular oscillation to the energy required to extract an electron, making possible a closer approach to resonance and therefore leading to a greater capture cross section. (This is the same effect as that causing broad absorption and emission spectra in polyatomic molecules.) Hydrogen would fall with the noble gases because its vibrational quantum energy,  $1/2$  ev, is so large. The importance of the resonance criterion in electron transfer to ions in this range of particle velocities has been emphasized recently by the beautiful work of Lindholm.

An energy dependence of  $W_{\alpha}$  in the high-energy portion is a question of much interest. The experimental status is complicated; and it is discussed in the following paper by Dr. Jesse. Theoretically, such an energy dependence must stem from change in the degradation spectra, and it therefore reflects the energy dependence of the relative cross sections for excitation and ionization, as averaged over both alpha-particle and electron spectra. We have inadequate information to anticipate, or to understand, whether the atomic and molecular gases differ in the energy dependence of  $W_{\alpha}$ , excluding the end of the track, and whether this  $W_{\alpha}$  should be equal to  $W_{\beta}$  or not. Indeed, as already emphasized, information on excitation cross sections for molecular gases is almost entirely lacking.

## 2. A Comparison of W Value for Alpha and Beta Particles; the Energy Dependence of W

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Some years ago a series of absolute W values was determined<sup>104</sup> for a large number of very pure gases for alpha particles emitted by Po<sup>210</sup> (5.3 Mev). More recently a series<sup>105</sup> of relative W values was determined for the same group of gases for an alpha particle reduced in energy to about 1 Mev by passage through sheets of mica. For such relative measurements argon was used as a standard gas. In addition relative ionization currents were also measured in an ionization chamber from beta-emitting sources of tritium, Ni<sup>63</sup>, and C<sup>14</sup>, the pressure being adjusted for each gas so that the total beta energy was absorbed. The  $W_\beta$  values for these gases, relative to argon as a standard, were found to be the same for the three beta sources used.

In comparing the  $W_\alpha$  and the relative  $W_\beta$  values, the purely arbitrary assumption is made that in argon  $W_\alpha/W_\beta = 1$  regardless of the energy of either the alpha or beta particles, within the ranges of energy so far investigated for either. Thus for all particle energies in argon  $W_\beta = W_\alpha = 26.4$  ev/ion pair, the absolute value determined for  $W_\alpha$  for Po alpha particles. This is a pure assumption, and its correctness or incorrectness can be verified only when absolute  $W_\beta$  measurements are available. Such measurements are considered later.

If with this assumption the  $W_\alpha$  values are plotted against the  $W_\beta$  values, the gases used are seen to be divided into two groups. For the noble gases and H<sub>2</sub> the points on the plot  $W_\alpha$  against  $W_\beta$  are seen to lie on a straight 45 degree line through the origin both for the Po alphas and the 1 Mev alphas. For all other gases the plotted points lie above this 45 degree line, showing a higher value for  $W_\alpha$  than for  $W_\beta$ . This means that in general in these latter gases the beta

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<sup>104</sup>W. P. Jesse and J. Sadauskis, Phys. Rev. 90, 1120 (1953).

<sup>105</sup>W. P. Jesse and J. Sadauskis, Phys. Rev. 97, 1668 (1955).

particle ionizes more efficiently than the alpha particle. Moreover, on the plot the  $W_\alpha$  values for the 1 Mev alphas are consistently higher than the  $W_\alpha$  values for Po alphas, indicating that  $W_\alpha/W_\beta$  is not constant with alpha energy in this group of gases but increases with decreasing alpha energy. It is tempting to conjecture that with increasing alpha energy the ratio  $W_\alpha/W_\beta$  in this group of gases decreases and approaches unity as a limit at very high alpha particle energies. It is interesting to note also that on the basis of the assumption that  $W_\alpha/W_\beta = 1$  for argon, one can predict an absolute  $W_\beta$  value for air of 34.0 ev/ion pair.

In order to test the assumption that in argon  $W_\alpha/W_\beta = 1$ , absolute measurements were made for  $W_\beta$  in a number of gases, the beta particles being those from a radioactive deposit of  $S^{35}$  on a very thin plastic film. The number of ion pairs produced per second was determined for each gas by measurements in an ionization chamber; the beta emission rate of the sample by proportional counting in a  $4\pi$  counter; the average energy of the beta particles by means of a graphical integration of the calculated beta ray spectral distribution curve. Two sets<sup>106, 107</sup> of absolute determinations of  $W_\beta$  for  $S^{35}$  were made. In the first the ionization measurements were corrected for the absorption of beta energy in the thin film and also for the interception of beta particles by the metal ring on which the film was mounted. In the second series a modification of technique eliminated the last correction.

The agreement between the two sets of determinations was good and led to an estimated value of  $W_\beta$  for air of 33.9 ev/ion pair. This is in excellent agreement with the value 34.0 ev/ion pair predicted as a result of the assumption that  $W_\alpha/W_\beta = 1$  in argon and also is in good agreement with absolute  $W_\beta$  values for air determined recently by quite different methods by other experimenters.<sup>108</sup> Explicitly then these experiments would indicate an equality in argon of  $W_\alpha$  for Po alphas and  $W_\beta$  for  $S^{35}$  betas within the limits of experimental error--about 1/2 per cent for  $W_\alpha$  and 1 per cent for  $W_\beta$ .

It was suggested that in the group of non-noble gases that the ratio  $W_\alpha/W_\beta$ , which is not a constant, approaches unity as a limit at

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<sup>106</sup>W. P. Jesse and J. Sadauskis, Phys. Rev. 107, 766 (1957).

<sup>107</sup>W. P. Jesse, Phys. Rev. 109, 2002 (1958).

<sup>108</sup>Bay, Mann, Seliger, and Wyckoff, Rad. Res. 7, 558 (1957).

very high alpha energies as  $W_\alpha$  diminishes. Accordingly very tentative measurements have been made to determine whether in the interval 5 to 9 Mev in alpha particle energy the mean  $W_\alpha$  for this interval is lower than the mean  $W_\alpha$  for the Po alpha and how closely it approaches the limiting  $W_\beta$  value. Very preliminary measurements made by comparing the difference in ionization in  $N_2$  and  $C_2H_4$  for Po alphas (5.3 Mev) and ThC' alphas (8.78 Mev) show a very distinct lowering of  $W_\alpha$  for this interval below the Po mean  $W$  value. The values for the interval, however, still seem about 0.5 ev/ion pair above the limiting  $W_\beta$  values. The uncertainty of these last results is large and the problem is being pursued further with more refined measurements.



### 3. Energy Loss per Ion Pair for Polonium Alpha Particles

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University of Basel, Switzerland

The interest that we originally had in knowing the value of  $W$  for different gases was a practical one. This quantity made possible the calculation of reaction  $Q$ -values based on charge measurements. It was clear to us from the beginning that a series of suppositions must be fulfilled, if these calculations are to be meaningful. Since the reactions investigated involved particles of different types and speeds than those of  $\text{Po-}\alpha$ , with which we exclusively determined the value of  $W$ , our calculations are permitted only insofar as  $W$  is independent of the type and speed of the particle. In the scope of our applications both suppositions prove to be permissible as a first approximation.

The method of slow ion collection is used for the determination of  $W$ . The preparation of  $\text{Po-}\alpha$  is deposited in a thin film from a  $\text{Ra-D-nitrate}$  solution onto a well polished nickel pin and attached to the negative plate of a parallel plate ionization chamber. The pressure of the gas under investigation is so chosen that the range of the particles is less than the distance between the plates.

The charge determination is made by induced calibration charges. At the connection between the collector electrode of the ionization chamber and the grid of the first tube a calibration condenser  $C_E$  of approximately  $1\ \mu\text{f}$  is attached. If the potential of point A (Figure 49) is varied by  $\Delta V$ , then the total capacity  $C_s$  from the grid to ground receives a voltage change of amount  $\Delta V \left( \frac{C_E}{C_s} \right)$ . This voltage change represents the true charge  $Q = \Delta V C_E$  on the grid.

A pulse of size  $\Delta V$  is generated when a current is switched through a precision resistance  $R_1$  ( $\sim 100\ \text{Ohms}$ ) by a mercury relay. The voltage across  $(R_1 + R_2)$  is made, with the help of a resistance  $R_3$ , equal to the voltage of a standard cell  $V_N$ . The equality of these two voltages is determined by a vibrating reed electrometer to an accuracy of  $10^{-4}$  v. The charge  $Q$  is

$$Q = \frac{R_1}{R_1 + R_2} V_N C_E$$

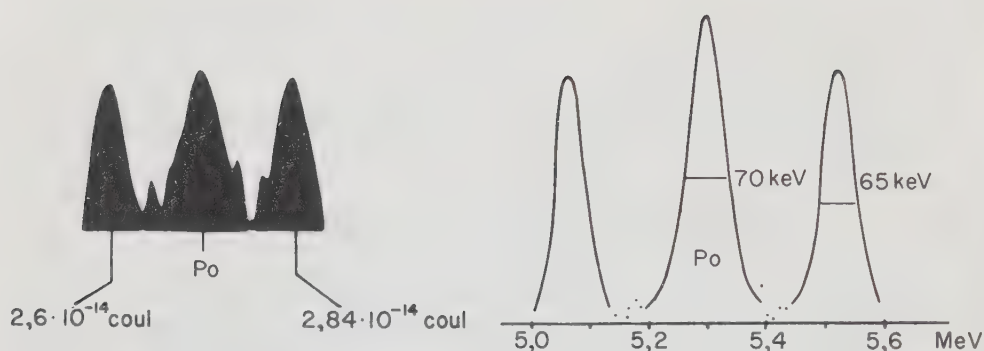


Figure 49. Measuring equipment used in the method of slow ion collection.

The calibration capacity  $C_E$  is determined with the aid of a standard condenser  $C_N$  to an accuracy of  $\pm 0.15$  per cent. This results in an error of  $\pm 0.2$  per cent for the calibrating charge. The pulses produced by the  $\alpha$ -particles are sorted according to number and size in a photographic pulse height analyzer.

Figure 50 shows an amplitude distribution of pulses from Po- $\alpha$ -particles in helium and Figure 51 from U- $\alpha$ -particles in nitrogen. Calibration charges are recorded simultaneously with those due to  $\alpha$ -particles, so that the latter can be determined by linear interpolation between the calibration marks. The mean error of the relation between the amplitudes of the  $\alpha$ -pulse and the calibration pulse amounts to about 0.1 per cent. Linearity and stability of the equipment were checked with calibration pulses.

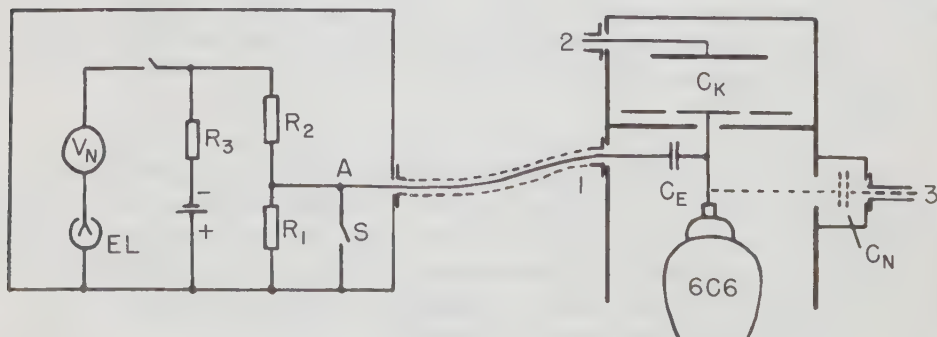


Figure 50. Amplitude distribution of pulses from Po- $\alpha$ -particles in helium.

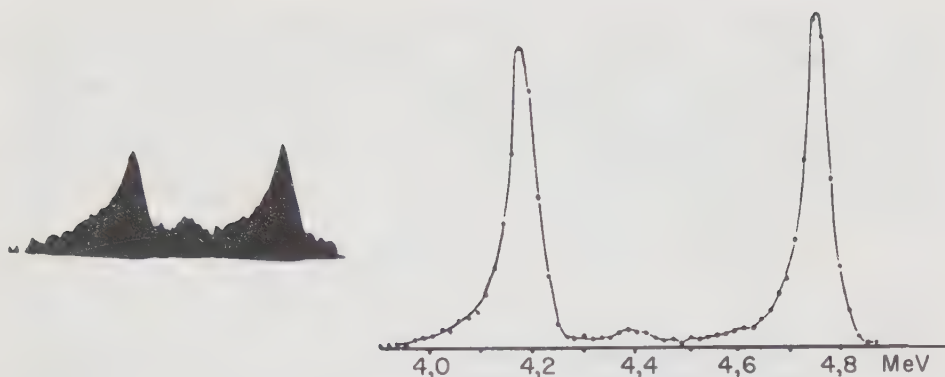


Figure 51. Amplitude distribution of pulses from U- $\alpha$ -particles in nitrogen.

The gases used were purified in some cases (methane,  $\text{CO}_2$ ,  $\text{BF}_3$ ,  $\text{H}_2\text{S}$ , butane and  $\text{SO}_2$ ). The purity was determined with the help of a mass spectrograph.

To determine the energy loss per ion pair  $W$  the number of ion pairs created and the energy of the  $\alpha$ -particle must be known. In order to be able to ascertain these values various corrections must be considered as follows:

- a. Noise in the measuring equipment.
- b. Energy loss by the  $\alpha$ -particles in the source.
- c. Recombination losses.
- d. Ballistic defect of the amplifier.

We will consider each of these corrections in turn.

Due to noise in the first amplifier tube the exact amplitude of the calibration charges is subject to a certain variation. This amplitude distribution can be represented well by a Gaussian curve whose half-value width with nitrogen gas in the ionization chamber is between 55 and 75 kev.

For the Po- $\alpha$ -source which we used the energy loss in the source can be neglected. The intensity of this source was about 70 particles per minute. Usable sources were prepared as follows: a highly polished, grease free nickel pin served as holder for the source. The pin was cooled with liquid air. At a distance of about 1 mm from the source holder a drop of diluted Ra-D-nitrate solution was held so that some of the solution condensed on the over surface of the pin.

We consider now the recombination losses. Part of the ions produced by the  $\alpha$ -particles in the gas are lost by recombination so that the measured charge  $Q$  is somewhat smaller than the charge  $Q_0$  which is actually produced. In our case of ionization by  $\alpha$ -particles the recombination losses can be computed by the theory of ionized columns developed by Jaffe. It furnishes the relation  $Q_0/Q$

$$\frac{Q_0}{Q} = 1 + \frac{\alpha N_0}{b u E \sin \phi} \frac{S(z)}{4\sqrt{2\pi}} \quad (1)$$

where

- $\alpha$  is the recombination coefficient
- $N_0$  is the number of ions per cm directly after the creation of the column
- $b$  is the radius of the column after its creation
- $u$  is the average mobility of the two ion types
- $E$  is the electric field strength
- $\phi$  is the angle between the axis of the column (track of the  $\alpha$ -particle) and the direction of the electric field.

The function  $S(z)$ , which takes consideration of the decrease in ion density through diffusion, increases with increasing  $z$  and has for  $z = \infty$  the value 1. The parameter  $z$  is defined as follows:

$$z = \frac{1}{2} \left( \frac{u}{Db} E \sin \phi \right)^2$$

where  $D$  is the diffusion coefficient. If  $E \sin \phi > 350$  V/cm, then  $S(z)$  equals 1 to a good approximation. According to equation (1) the charge collected on the electrodes depends on the angle between the  $\alpha$ -particle track and the direction of the field. For ion collection with  $S(z) = 1$  with a given pressure and field strength

$$\frac{Q_0}{Q} = 1 + \frac{R}{\sin \phi}$$

The constant  $R$  represents the saturation defect relative to the measured charge  $Q$  for  $\phi = 90^\circ$ , i. e., for  $\alpha$ -particles that move perpendicularly to the electric field. Since the number of  $\alpha$ -particles per unit angle varies as  $dn = N_0 \sin \phi d\phi$  the charge distribution resulting from the recombination at various angles is calculated to be

$$\frac{dn}{dQ} = \frac{N_0 R^2}{Q_0 (1 - Q/Q_0)^2 \sqrt{(1 - Q_0/Q)^2 - R^2}}$$

Figure 52 shows as an example a pulse spectrum which demonstrates the smearing effect of recombination for oxygen at 6 atm with a field strength of 2.75 kv/cm. The solid curve shows the spectrum calculated by equation (2) with  $R = 0.24$  without considerations of noise.

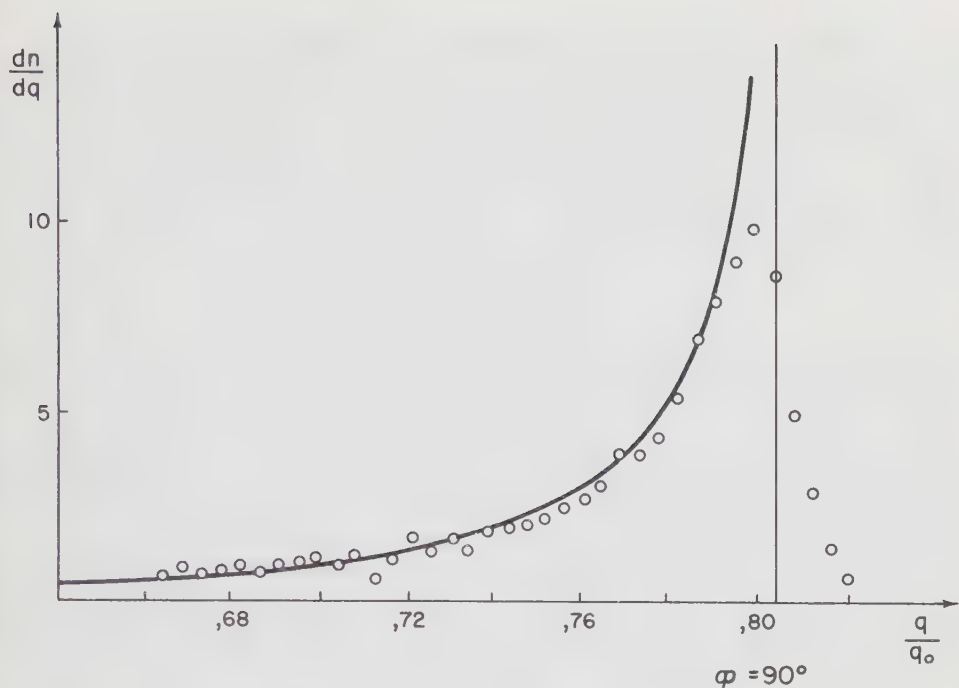


Figure 52. Example of a pulse spectrum demonstrating the smearing effect of recombination for oxygen.

For the determination of  $W$  the knowledge of the saturation charge is essential. According to equation (1)

$$\frac{Q_0}{Q} = 1 + \frac{R(p)}{E}$$

The quantity  $R = \frac{a N_0}{b u \sin \phi} \frac{S(z)}{4\sqrt{2\pi}}$  can be considered as a constant for constant pressure, definite angle between particle track and field direction, and field strength that is not too small. The saturation charges  $Q_0$ , therefore, may be determined by extrapolation for  $1/E \rightarrow 0$  of the saturation line

$$\frac{1}{Q} = \frac{1}{Q_0} \left( 1 + \frac{R(p)}{E} \right) \quad (3)$$

From this assumption it is also expected that the saturation lines, which one receives from equation (3) by varying the pressure, cut the ordinate axis at a common point which is independent of pressure. This relationship is most easily checked by plotting the reciprocal of the measured charge vs. the reciprocal of the field strength.  $1/Q_0$  is obtained by a linear extrapolation of  $1/E \rightarrow 0$ .



The last of the four corrections is the ballistic defect of the amplifier. In measuring the charge by means of a pulse amplifier the amplitude of the output pulse is a measure of the charge pulse at the input. If the amplifier receives a charge in the form of a  $\delta$ -function, then the output signal has an amplitude  $A_0$  which depends only on the amplification factor. For an input signal of longer duration but of equal charge  $Q$  the output amplitude  $A$  is somewhat smaller:

$$A = A_0 (1 - b)$$

The quantity  $b$ , designated as ballistic defect, depends on the shape and duration of the input pulse and on the frequency response of the amplifier. The ballistic defect is small, if the duration of the input pulse is short compared with the time constant of the amplifier. Figure 53 shows the influence of the ballistic defect on the measurement of a saturation curve. Curves (a) and (b) were taken with two different time constants. As expected it is seen that by reducing the duration of the current pulse the measured charge is reduced. The ballistic defect can be calculated and the correction determined. The corrected curve is likewise shown in Figure 53 and it shows the result expected from the assumptions made in Jaffé's theory.

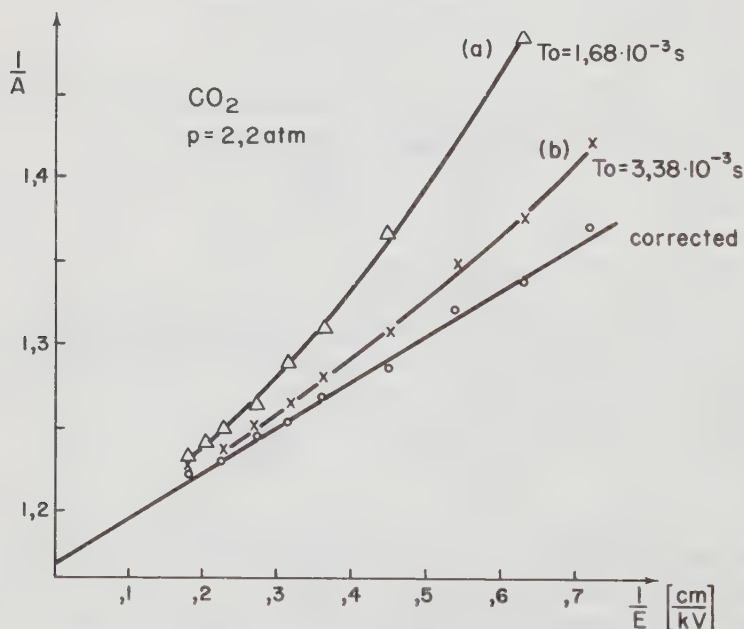


Figure 53. Influence of the ballistic defect on the measurement of a saturation curve.

For gases with little electron affinity the ballistic defect has no influence on the results of the saturation curve when the source is attached to the negative electrode of the ionization chamber. The most probable amplitude is associated with tracks which are perpendicular to the field, so that only the electrons contribute to the current, a very short pulse results, and hence, practically no ballistic defect occurs. It is experimentally confirmed that by decreasing the time constant in this case the amplitude distribution merely becomes broader but its maximum is unchanged.

Saturation curves of 15 different gases for different pressures and for field strengths up to 11 kv/cm were taken. All measurements were corrected for ballistic defect. The results show with the exception of two gases, namely nitrogen and carbon dioxide, the outcome expected from Jaffé's theory. Examples of this behavior are shown in Figures 54, 55 and 56. Figure 57 shows the special case of nitrogen. At high field strengths the saturation curves show a region of linear dependence which one can use for extrapolation. The saturation charges as determined proved to be pressure independent to within 0.2 per cent.

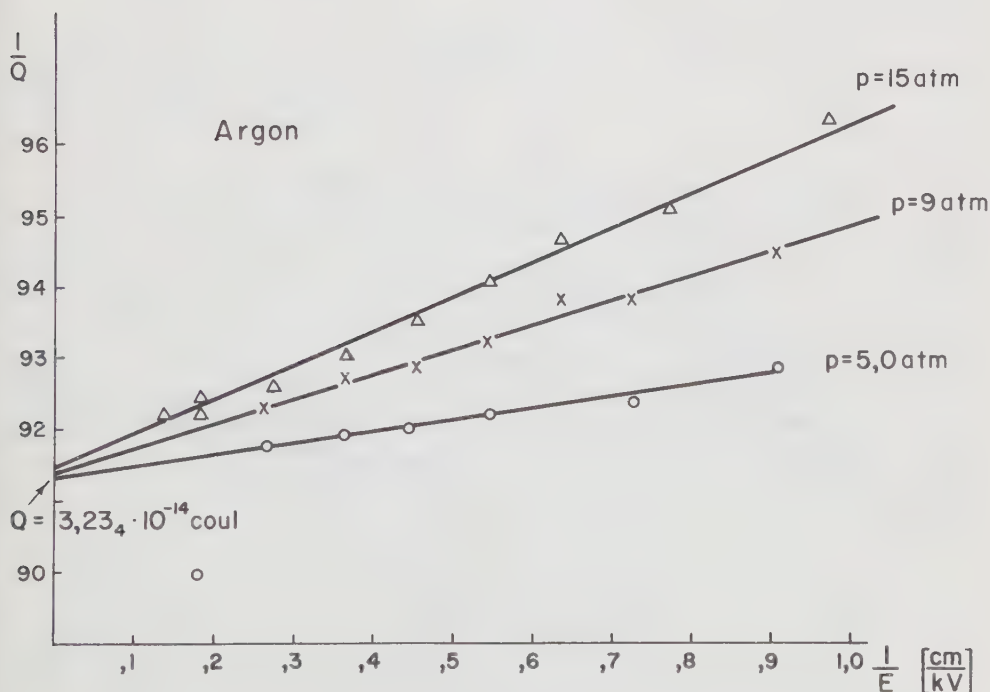


Figure 54. Saturation curves as a function of pressure and field for argon.

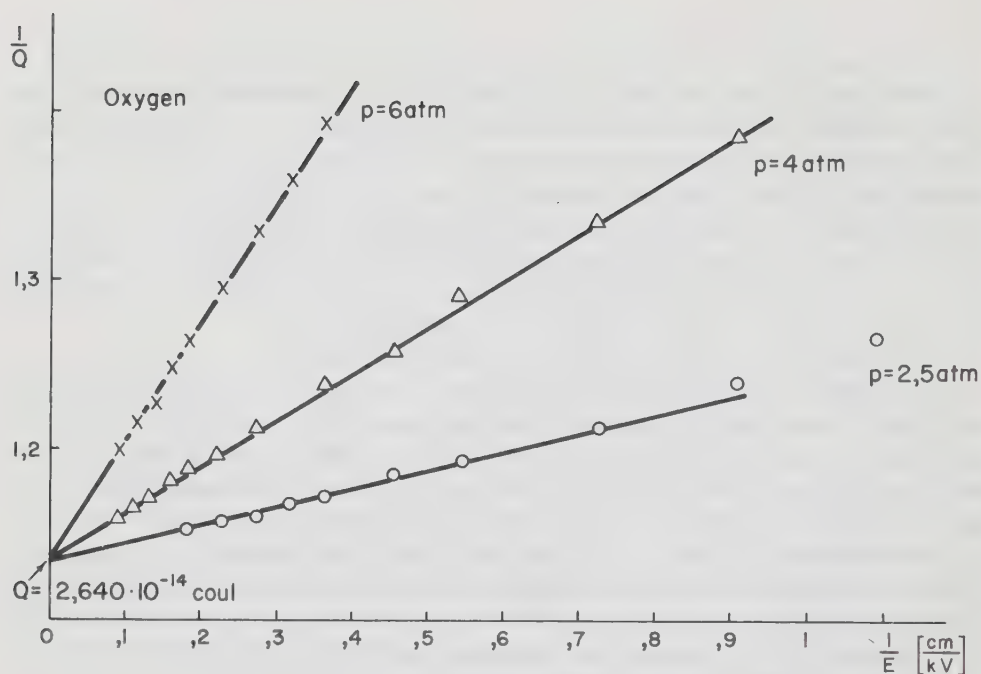


Figure 55. Saturation curves as a function of pressure and field for oxygen.

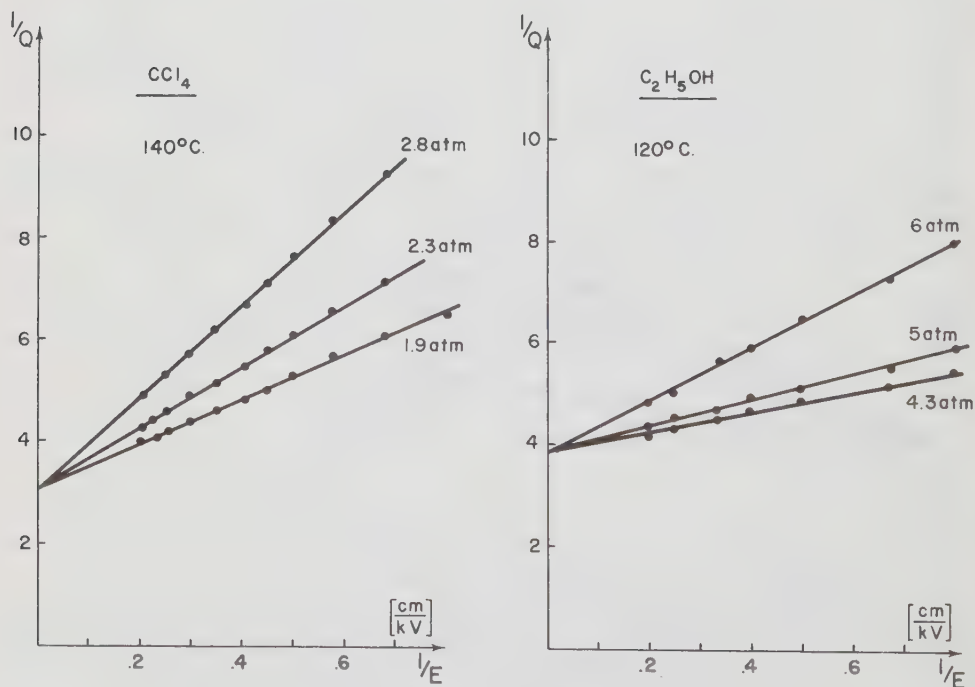


Figure 56. Saturation curves as a function of pressure and field for  $\text{CCl}_4$  and  $\text{C}_2\text{H}_5\text{OH}$ .

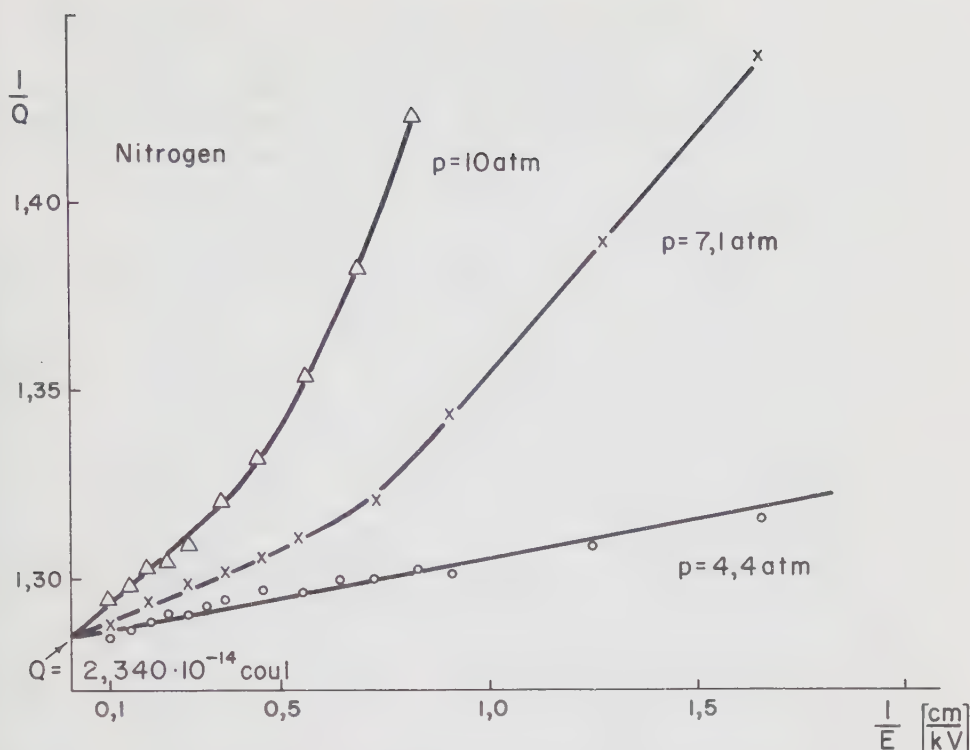


Figure 57. Saturation curves as a function of pressure and field for nitrogen.

Of the gases investigated  $\text{CO}_2$  shows a special anomaly. This is shown in Figure 58 where the saturation curves are plotted as a function of the reciprocal electric field in the top set of curves and as a function of the reciprocal electron drift velocity in the bottom set of curves. The saturation curves are dependent. With increasing pressure a systematic increase of the extrapolated charge for  $1/E \rightarrow 0$  is observed. A searching investigation of this dependence leads to the following conclusion: in the region of values for  $E/p$  investigated the electron mobility  $u$  is not constant for  $\text{CO}_2$ . If the anomalous pressure dependence really originated from the field strength dependence of  $u$ , then  $1/Q$  must be a function not of  $1/E$  but of  $1/uE = 1/v$ , where the symbol  $v$  stands for the electron drift velocity. In plotting the curves of Figure 58b  $1/v$  is taken as the abscissa rather than  $1/E$ . In this presentation the pressure dependence of the saturation charge vanishes.

With the saturation charge so determined the energy loss per ion pair  $W$  can be obtained. The results using the Po- $\alpha$ -particles

are shown in Table XIII where the results of measurements made by various observers are compared. The table shows that for all gases except helium the values of  $W$  are consistent within  $\pm 0.5$  ev. The large variation in the case of helium is caused by the presence of impurities in helium, as shown by Jesse and Sadauskis.

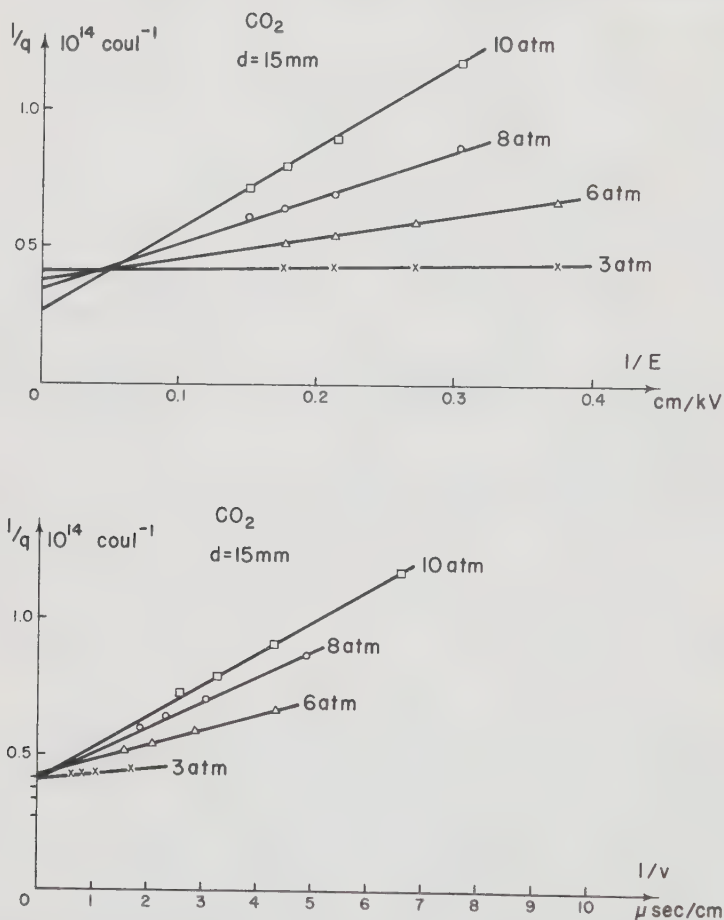


Figure 58. Saturation curves as a function of pressure for  $\text{CO}_2$  plotted (a) with respect to reciprocal field and (b) with respect to reciprocal electron drift velocity.



TABLE XIII

Measurements of W

| Gas                              | Jesse and<br>Sadauskis<br>(1953) | Sharpe<br>(1952) | Valentine<br>and<br>Curran<br>(1952) | Huber and<br>Co-workers*<br>(1953, 1955) | Bortner<br>and<br>Hurst<br>(1954) |
|----------------------------------|----------------------------------|------------------|--------------------------------------|------------------------------------------|-----------------------------------|
| A                                | 26.4 ev                          | 26.3 ev          | 25.9 ev                              | 26.25 ev                                 | 26.4 ev                           |
| He                               | 42.7                             | -                | 31.7                                 | 29.6                                     | 46.0                              |
| Ne                               | 36.8                             | -                | -                                    | -                                        | -                                 |
| Kr                               | 24.1                             | -                | -                                    | -                                        | -                                 |
| Xe                               | 21.9                             | -                | -                                    | -                                        | -                                 |
| H <sub>2</sub>                   | 36.3                             | -                | 37.0                                 | 35.96                                    | 37.0                              |
| CO <sub>2</sub>                  | 34.5                             | 34.2             | -                                    | 34.3                                     | 34.3                              |
| Air                              | 35.5                             | 35.6             | 35.2                                 | 34.95                                    | 35.0                              |
| O <sub>2</sub>                   | 32.5                             | 32.9             | 32.2                                 | 32.17                                    | 32.2                              |
| N <sub>2</sub>                   | 36.6                             | 36.4             | 36.0                                 | 36.5                                     | 36.3                              |
| CH <sub>4</sub>                  | 29.2                             | 29.1             | 29.0                                 | 29.0                                     | 29.4                              |
| C <sub>2</sub> H <sub>2</sub>    | 27.5                             | -                | -                                    | -                                        | -                                 |
| C <sub>2</sub> H <sub>6</sub>    | 26.6                             | -                | -                                    | -                                        | -                                 |
| C <sub>2</sub> H <sub>4</sub>    | 28.4                             | -                | -                                    | -                                        | 28.0                              |
| BF <sub>3</sub>                  | -                                | -                | -                                    | 35.3                                     | 36.0                              |
| SF <sub>6</sub>                  | -                                | -                | -                                    | -                                        | 35.7                              |
| Freon 12                         | -                                | -                | -                                    | -                                        | 29.5                              |
| H <sub>2</sub> S                 | -                                | -                | -                                    | 23.4                                     | -                                 |
| NH <sub>3</sub>                  | -                                | -                | -                                    | 30.5                                     | -                                 |
| C <sub>4</sub> H <sub>10</sub>   | -                                | -                | -                                    | 23.0                                     | -                                 |
| SO <sub>2</sub>                  | -                                | -                | -                                    | 32.5                                     | -                                 |
| C Cl <sub>4</sub>                | -                                | -                | -                                    | 25.9                                     | -                                 |
| C <sub>2</sub> H <sub>5</sub> OH | -                                | -                | -                                    | 32.6                                     | -                                 |

\*Biber, Huber and Müller (1955) and Haeberli, Huber and Baldinger (1953).

Editor's Note: A portion of this paper is contained in F. Widder and T. Huber, *Helv. Phys. Acta* 31, 601 (1958).

#### 4. Alpha Particle Ionization of Binary Gas Mixtures

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and

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This report deals mainly with a summary of the work done at ORNL on the question of the amount of ionization produced by alpha particles losing their energy in mixtures of two gases. No effort will be made to present a general summary of the work at other laboratories, although some of this work will be quoted when a comparison of results seems advisable. The material already published <sup>109</sup>, <sup>110</sup>, <sup>111</sup> will be covered in an abbreviated fashion in order to save space for more recent unpublished work.

For purposes of discussion it will be useful to distinguish between two general effects on the amount of ionization produced by alpha particles in gas mixtures. When very small amounts of some gas are added to certain other gases, particularly the noble gases, marked increases in the total amount of ionization are observed. These increases in observed ionization are explained in terms of the transfer of excitation energy from the main gas component to the minor gas component. When the energy level of the excited states, and in most cases these are metastable states, exceeds the ionization potential of the minor component, ionization of the latter can take place, accounting for the increased amount of total ionization. The

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<sup>109</sup>T. E. Bortner and G. S. Hurst, Phys. Rev. 93, 1236 (1954).

<sup>110</sup>C. E. Melton, G. S. Hurst, and T. E. Bortner, Phys. Rev. 96, 643 (1954).

<sup>111</sup>H. J. Moe, T. E. Bortner, and G. S. Hurst, J. Phys. Chem. 61, 422 (1957).

effect has been referred to as the Jesse effect<sup>112</sup> in honor of W. P. Jesse who first called attention to its importance in the case of alpha particle ionization of He.<sup>113, 114, 115</sup>

The second effect takes place more gradually as the two components of the mixture assume comparable proportions. In cases when the Jesse effect is not appreciable, the value of W (average amount of energy required to create an ion pair) of any mixture of the gases usually lies between the extreme values and changes smoothly from one extreme to the other as the composition of the mixture is changed. This effect has been studied by the Swiss group<sup>116, 117</sup> and more recently at this Laboratory.<sup>109, 111</sup> In these studies it has been found that the value of W for a mixture of two gases in arbitrary proportions can be expressed in terms of the W values for the pure gases and an empirical constant. Each set of two gases has a characteristic empirical constant, but their magnitudes are not simply related to known constants for the gases. In spite of the fact that the empirical constant is not quantitatively understood, gas mixtures behaving in this way will be referred to as "regular" mixtures.

Figure 59 shows the values of W obtained for mixtures of H<sub>2</sub>, N<sub>2</sub>, CH<sub>4</sub>, and A with He. Even when less than one per cent of H<sub>2</sub>, N<sub>2</sub>, CH<sub>4</sub>, or A is added to He, the value of W drops from 46 (ev per ion pair) to approximately 30. This is consistent with the results of Jesse who shows that a few hundred parts per million of various

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<sup>112</sup>R. L. Platzman, "The Physical and Chemical Basis of Mechanisms in Radiation Biology," in Radiation Biology & Medicine (Walter D. Claus, ed.), pp. 15-72 (1958), Addison-Wesley Publishing Co., Inc., Reading, Mass.

<sup>113</sup>W. P. Jesse and J. Sadauskis, Phys. Rev. 77, 417 (1950).

<sup>114</sup>Ibid., 90, 1120 (1953).

<sup>115</sup>Ibid., 100, 1755 (1955).

<sup>116</sup>Huber, Baldinger, and Haeberli, Helv. Phys. Acta. 23, Suppl. 111 (1949).

<sup>117</sup>Haeberli, Huber, and Baldinger, Helv. Phys. Acta. 26, 145 (1953).

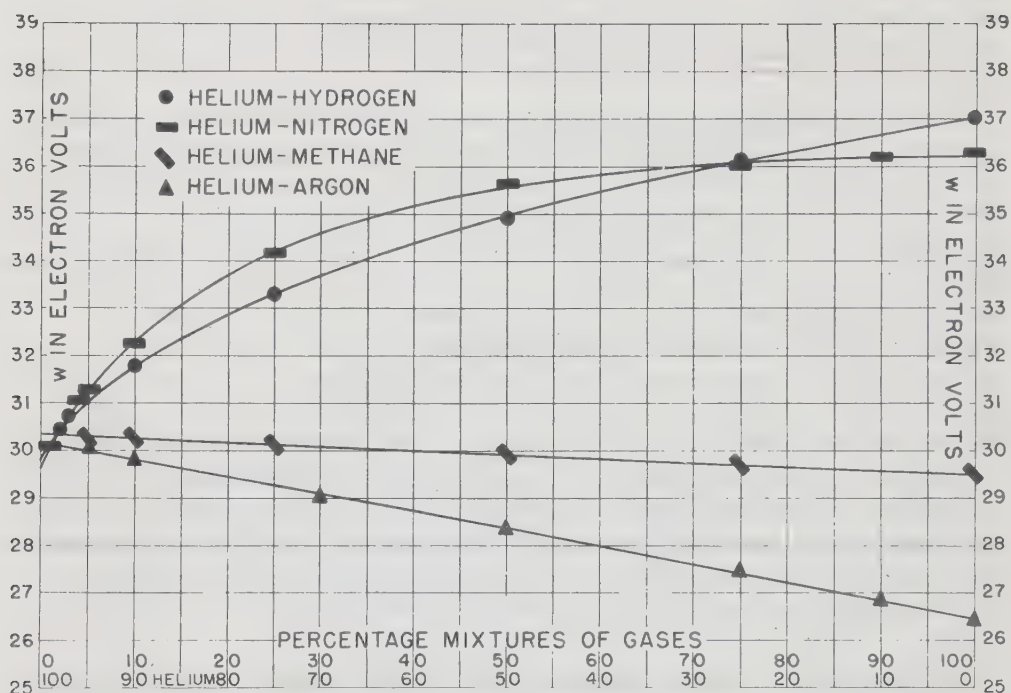


Figure 59.  $W$  for mixtures of He with  $H_2$ ,  $N_2$ ,  $CH_4$  and A.  $W$  for pure He is 46.0.

gases is sufficient to discharge most of the metastable states which are left excited in He as a result of alpha particle ionization. Actually, as Figure 59 shows, the low value of  $W$  observed when various gases are added as small percentages depends slightly on the kind of gas which is added to He. This effect has been explained by Platzman<sup>118</sup> in terms of "subexcitation electrons." As He is further diluted with the other gases, the value of  $W$  changes smoothly to the  $W$  value of the second component. This effect is similar to the effect in "regular mixtures" and will be discussed in a later section.

A systematic study<sup>110</sup> of the alpha particle ionization of argon mixed with various gases has shown a number of interesting effects. Figure 60 is an illustration of the type of results obtained. The lower curve shows the behavior of  $W$  when argon is mixed with acetylene. The ionization potential of acetylene is near but probably

<sup>118</sup>R. L. Platzman, Rad. Res. 2, 1 (1955).

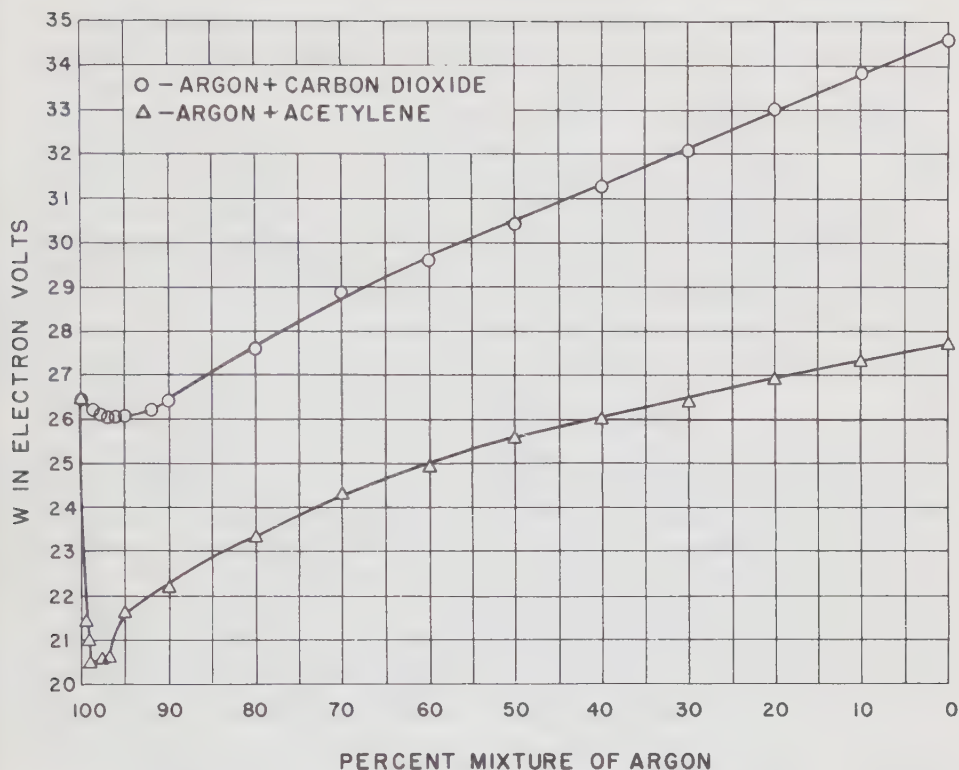


Figure 60. W values for Pu alpha particles in mixtures of A + CO<sub>2</sub> and A + CH<sub>4</sub>.

slightly less than the metastable state in argon, thus the pronounced decrease in W is explained in precisely the same way as in the Jesse effect in He. The upper curve for argon plus carbon dioxide shows an increase in ionization even though the ionization potential of CO<sub>2</sub> is definitely higher than the metastable level in A. The results for mixtures of several gases with A are summarized in Figure 61 where the minimum W values are plotted as a function of ionization potential of the added gases. The numbers in parentheses are the percentages of the added gases at which the minimum W values occur. The cases where the ionization potential of the impurity is greater than the metastable states in A cannot be understood in terms of the Jesse effect. A similar process, i.e., the transfer of energy from excited states in A at about 15 ev, may be involved. Since the explanation is not entirely clear, it will be referred to as the anomalous effect in A.



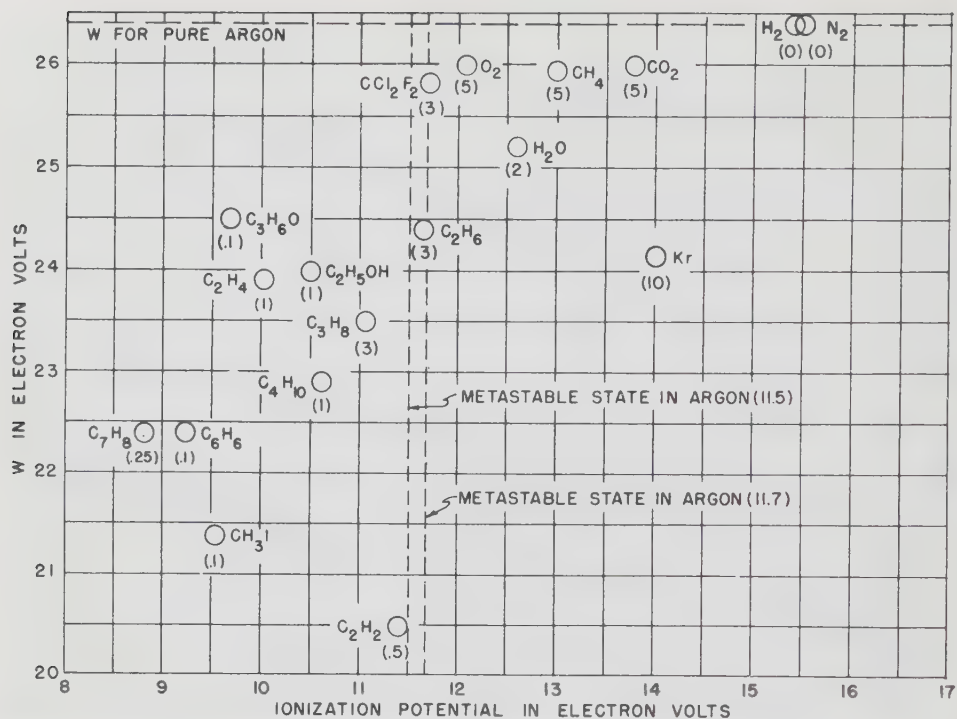


Figure 61. Minimum values of  $W$  for argon plus impurities as a function of ionization potentials of impurities. Numbers in parenthesis are per cents of impurities at which the minimum values are obtained.

For a large number of binary gas mixtures it has been found that the  $W$  for a mixture,  $W_{ij}$ , is given by

$$\frac{1}{W_{ij}} = \left( \frac{1}{W_i} - \frac{1}{W_j} \right) Z_{ij} + \frac{1}{W_j} \quad (1)$$

where

$$Z_{ij} = \frac{P_i}{P_i + a_{ij} P_j} \quad (2)$$

$W_i$  is the value for the gas having partial pressure  $P_i$ ,  $W_j$  is the  $W$  value for the gas having partial pressure  $P_j$ , and  $a_{ij}$  is an empirical constant for a particular mixture. This extremely simple empirical formula has the same form as the Huber et al.<sup>116</sup> equation. Specifically, if we replace  $Z_{ij}$  with

$$Z'_{ij} = \frac{S_i P_i}{S_i P_i + S_j P_j} \quad (3)$$

where  $S_i$  and  $S_j$  are molecular stopping powers of the gases for alpha particles, we have their results.

Equations (1) and (3) involve the over-simplified model that the amount of ionization in a binary mixture is simply equal to the sum of the ionization in the two components in their pure forms. If this were true the value of  $a_{ij}$  in equation (2) would simply be  $S_j/S_i$ . It has been shown that the value of  $a_{ij}$  may differ from  $S_j/S_i$  by large factors and this is not surprising considering the gross oversimplification involved. In spite of the fact that  $a_{ij}$  is very poorly correlated with  $S_j/S_i$ , equations (1) and (2) represent the experimental results for  $W_{ij}$  with good accuracy.

Figure 62 shows the results of  $H_2$ -A as an example. When  $1/W_{ij}$  is plotted against  $Z'_{ij}$ , the results deviate from a straight line, but when plotted against  $Z_{ij}$  they fall very close to a straight line. In this example  $S_j/S_i = 4.5$  while the best fit is obtained when  $a_{ij} = 1.8$ . Table XIV shows several other cases where  $a_{ij}$  can be compared to  $S_j/S_i$ . For the cases where He is one of the gases in the mixture, the W values for "contaminated" He were used (see Figure 59). For A mixtures only the cases A- $H_2$  and A- $N_2$  are included since these are the only two cases not complicated by the anomalous effect.

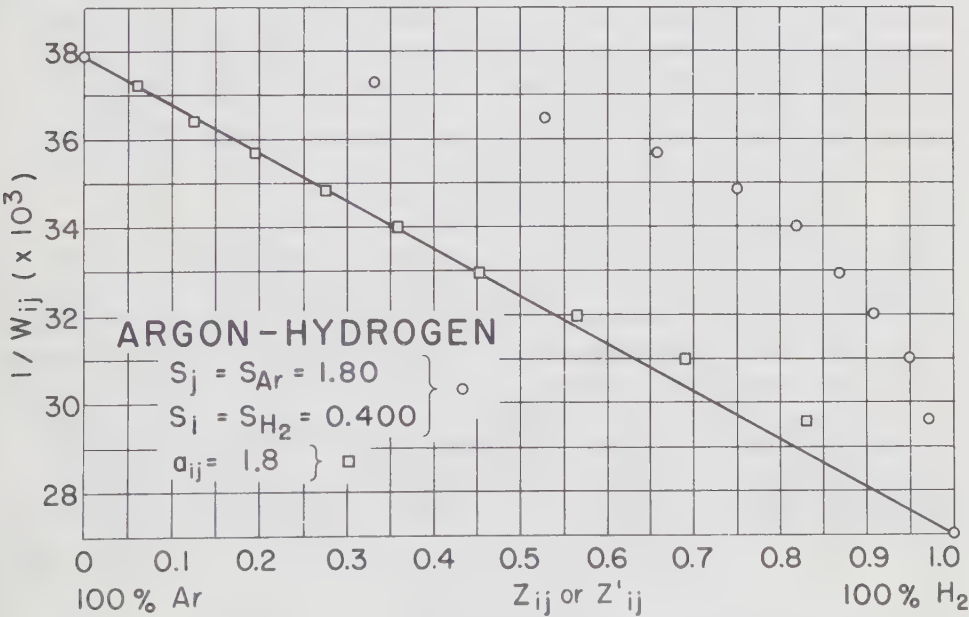


Figure 62.  $1/W_{ij}$  as a function of  $Z_{ij}$  or  $Z'_{ij}$  for A -  $H_2$  mixture.

TABLE XIV

Ionization in Binary Gas Mixtures -  
 Constants Required for Evaluation of Equations (1), (2)

| Mixture                       |                   | $W_i$ | $W_j$ | $a_{ij}$ | $S_j/S_i$ | Ref |
|-------------------------------|-------------------|-------|-------|----------|-----------|-----|
| (i)                           | (j)               |       |       |          |           |     |
| N <sub>2</sub>                | - H <sub>2</sub>  | 36.3  | 37.0  | 0.28     | 0.21      | 109 |
| N <sub>2</sub>                | - A               | 36.3  | 26.4  | 0.53     | 0.95      | 109 |
| N <sub>2</sub>                | - O <sub>2</sub>  | 36.3  | 32.2  | 1.06     | 1.06      | 109 |
| He                            | - A               | 30.1  | 26.4  | 0.75     | 5.85      | 109 |
| He                            | - H <sub>2</sub>  | 29.7  | 37.0  | 3.55     | 1.30      | 109 |
| He                            | - N <sub>2</sub>  | 29.7  | 36.3  | 8.47     | 6.10      | 109 |
| He                            | - CH <sub>4</sub> | 30.3  | 29.4  | 0.68     | 5.24      | 109 |
| H <sub>2</sub>                | - A               | 37.0  | 26.4  | 1.78     | 4.5       | 109 |
| H <sub>2</sub>                | - CH <sub>4</sub> | 37.0  | 29.4  | 4.03     | 4.03      | 109 |
| C <sub>2</sub> H <sub>2</sub> | - N <sub>2</sub>  | 27.8  | 36.3  | 0.26     | 0.88      | 111 |
| C <sub>2</sub> H <sub>2</sub> | - CO <sub>2</sub> | 27.8  | 34.3  | 0.93     | 1.34      | 111 |
| C <sub>2</sub> H <sub>2</sub> | - CH <sub>4</sub> | 27.8  | 29.4  | 0.39     | 0.78      | 111 |
| CH <sub>4</sub>               | - N <sub>2</sub>  | 39.4  | 36.3  | 0.62     | 1.13      | 111 |
| C <sub>2</sub> H <sub>2</sub> | - He              | 27.8  | 30.3  | 0.058    | 0.18      | 111 |

In order to study the relationship between the  $a_{ij}$  values of a series of gases, it is convenient to replace  $a_{ij}$  in equation (2) by  $W_j f_j / W_i f_i$  in which case equation (1) can be rewritten in the form

$$W_{ij} = (W_i - W_j) Z''_{ij} + W_j \quad (4)$$

where

$$Z''_{ij} = \frac{f_i P_i}{f_i P_i + f_j P_j} - \frac{P_i}{P_i + f_j / f_i P_j} \quad (5)$$

This can be plotted as a linear relationship between  $W_{ij}$  and  $Z_{ij}''$  provided the proper value of the empirical constant  $f_j/f_i$  is used for any pair of gases. It is interesting to determine whether constants  $f_i$  and  $f_j$  can be uniquely associated with gases  $i$  and  $j$  such that equations (4) and (5) predict the measured values of  $W$ .

For this purpose, measurements were made of  $W_{ij}$  for all possible combinations (in pairs) of the following gases:  $H_2$ ,  $N_2$ ,  $CO_2$ ,  $CH_4$ ,  $C_2H_4$ , and  $C_3H_6$  (cyclopropane). These were chosen to represent a rather wide range of  $W$  values and did not include any of the noble gases where the complicating factors of the Jesse effect and the anomalous effect are present. For each pair of these gases (with the exception of  $N_2-H_2$  in which the  $W$ 's are too close together) a constant  $f_j/f_i$  was determined such as to give the best straight line using equation (4). These constants are listed in Column 2 of Table XV. The  $W$  values were all measured in comparison with nitrogen whose  $W$  was taken as 36.3 ev/ip. With the ionization chamber described earlier, <sup>109</sup> comparative  $W$  measurements are reproducible to about 0.2 per cent. The experimental values listed in Column 2 of Table XV have been assigned a probable error. These were obtained by finding that variation in  $f_f/f_i$  which would move the  $W$  value at the mid point of the curve ( $Z_{ij}'' = 0.5$ ) off the line by as much as 0.2 per cent. These probable errors are therefore seen to be relatively large in the case of mixtures whose two component gases have  $W$ 's which are close together. In these cases, large changes in  $f_j/f_i$  have little effect in shifting the curve from a straight line. The set of ratios shown in Column 3 of Table XV was chosen to satisfy the following conditions: (1) the individual ratios satisfy the relationship

$$f_j/f_i = \frac{f_j/f_k}{f_i/f_k} \quad (6)$$

where,  $i$ ,  $j$ , and  $k$  designate any three gases; and (2) the set of values must be compatible with the experimental data. In Column 3, Table XV condition (1) was rigorously applied. The extent to which condition (2) holds may be judged from Figure 63. In this figure  $W_{ij}$  is plotted against  $Z_{ij}''$  (equation 5) where the values of  $f_j/f_i$  are taken directly from Column 3, Table II.

Since the numbers in Column 2 of Table XV are in reasonable agreement with those in Column 3, it is possible to write for the  $a_{ij}$  values of these gases

$$a_{ij} = \frac{a_{ik}}{a_{jk}} \quad (7)$$

Generality of this result is not claimed, but such a relationship may hold for many other "regular" gas mixtures, especially in those

cases where the W values for the two components in the mixture are appreciably different.

The authors are pleased to acknowledge the help of T. E. Bortner, ORNL, and J. E. Purcell, Berea College, who made many suggestions and carried out much of the experimental work reported in this article.

TABLE XV

Ionization in Binary Mixtures of Regular Gases -  
Ratio  $f_j/f_i$  Appearing in Equations (4) and (5)

| Mixture                       |                                 | $f_j/f_i$<br>(measured) | $f_j/f_i$<br>(calculated) |
|-------------------------------|---------------------------------|-------------------------|---------------------------|
| (i)                           | (j)                             |                         |                           |
| N <sub>2</sub>                | - CH <sub>4</sub>               | 1.65 ± 0.06             | 1.8                       |
| N <sub>2</sub>                | - CO <sub>2</sub>               | 1.6 ± 0.2               | 1.8                       |
| N <sub>2</sub>                | - C <sub>2</sub> H <sub>4</sub> | 3.2 ± 0.1               | 3.4                       |
| N <sub>2</sub>                | - C <sub>3</sub> H <sub>6</sub> | 4.50 ± 0.14             | 4.5                       |
| CO <sub>2</sub>               | - CH <sub>4</sub>               | 1.50 ± 0.06             | 1.0                       |
| CO <sub>2</sub>               | - C <sub>2</sub> H <sub>4</sub> | 1.67 ± 0.05             | 1.88                      |
| CO <sub>2</sub>               | - C <sub>3</sub> H <sub>6</sub> | 2.5 ± 0.1               | 2.5                       |
| CH <sub>4</sub>               | - C <sub>2</sub> H <sub>4</sub> | 2.0 ± 0.5               | 1.88                      |
| CH <sub>4</sub>               | - C <sub>3</sub> H <sub>6</sub> | 2.3 ± 0.2               | 2.5                       |
| C <sub>2</sub> H <sub>4</sub> | - C <sub>3</sub> H <sub>6</sub> | 1.3 ± 0.15              | 1.36                      |
| H <sub>2</sub>                | - CO <sub>2</sub>               | 3.5 ± 0.9               | 3.6                       |
| H <sub>2</sub>                | - CH <sub>4</sub>               | 3.8 ± 0.15              | 3.6                       |
| H <sub>2</sub>                | - C <sub>2</sub> H <sub>4</sub> | 7.2 ± 0.2               | 6.8                       |
| H <sub>2</sub>                | - C <sub>3</sub> H <sub>6</sub> | 9.0 ± 0.25              | 9.0                       |



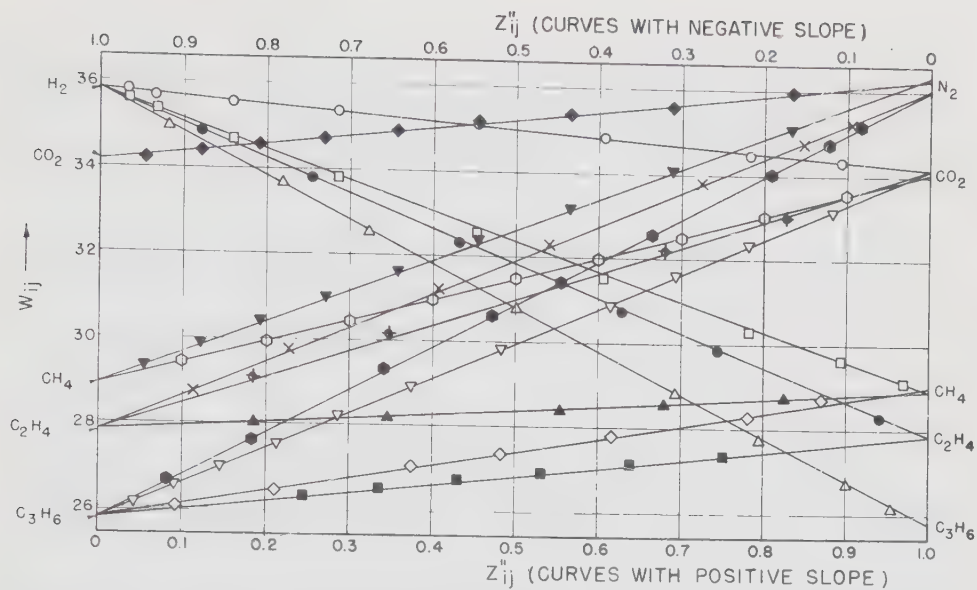


Figure 63.  $W_{ij}$  as a function of  $Z_{ij}''$  for mixtures of regular gases (f ratios from Column 3, Table XV are used in the computation of  $Z_{ij}''$ ).

## VI. MICROSCOPIC AND MACROSCOPIC ENERGY LOSS DISTRIBUTIONS

### 1. Theoretical Reviews: A Summary

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National Bureau of Standards

These sessions dealt primarily with spectral and angular distributions of energy losses of electrons through elementary processes in condensed materials. The localization and spatial distribution of energy losses within a material were not discussed-- basically because of lack of information. Subsequent effects of energy losses were only touched upon in the description, by Ferrell, of a possible mechanism of energy escape as electromagnetic radiation.

The direct evidence on energy losses derives from observations of the energy and angular distribution of electrons that emerge from exceedingly thin foils (100-1000 Å). The preparation of these foils is so difficult that their microscopic homogeneity and actual thickness are still poorly controlled. Therefore only relative, and hardly any absolute, experimental data are available on the probability of a given energy loss and deflection per unit pathlength of an electron traversing a given material. It is particularly important to check clearly whether the probability of multiple characteristic energy losses,  $E_c$ ,  $2E_c$ ,  $3E_c$ , ...  $nE_c$ , in a foil is given by a Poisson distribution  $\exp(-m)m^n/n!$ , whose parameter  $m$  is proportional to the foil thickness. There has been only one study of this type leading to an absolute, though rather crude, determination of the collision probability.<sup>119</sup> Theory, on its side, accomplishes at this time little more than framing the experimental observations within a rather formal description of the absorption of energy by condensed matter. Thus much progress of both experiment and theory is required before this field of physics emerges from its infancy.

<sup>119</sup>A. W. Blackstock, R. H. Ritchie, and D. Birkhoff, Phys. Rev. 100, 1078 (1955).

The subject was introduced by two reviews of general theory, by Fano and by Nozières, which were very similar in spirit and dealt with the passage of electrons through infinite, homogeneous, isotropic media, disregarding the effects of boundaries and inhomogeneities. Both reviews stressed that: (a) the probability of an elementary inelastic process with a given energy loss and deflection, per unit path of an electron in a given material, is conveniently expressed in terms of a generalized dielectric constant  $E(k, \omega)$  of the material; (b) the great majority of these collisions results from a longitudinal interaction of the incident electron with the electrons of the material; (c) the energy levels of the material excited by the longitudinal interaction are influenced greatly by the Coulomb interaction between charges within distant atoms, whereas the levels excited by transverse interaction (e.g., with optical radiation) are not; (d) therefore the spectra of longitudinal and transverse excitations are radically different; (e) nevertheless the same dielectric constant enters the theories of electron energy losses and of other (e.g., optical) phenomena, so that the spectra of energy loss observed in a material can be related to other observable properties of the same material; (f) the function  $E(k, \omega)$  of a material may be defined, according to Van Hove, as a Fourier component of the autocorrelation between the positions of its atomic electrons. Fano stressed particularly that, to fulfill sum rules and dispersion relations, the spectrum of energy losses in any condensed material must have its major peak above the range of optical absorption. Nozières pointed out that also the incoherent scattering function of a material (and therefore the Compton effect probability) and the Coulomb interaction energy of its electrons in the ground state can be expressed in terms of  $E(k, \omega)$  when the optical electrons of a metal are treated schematically as an electron gas. Some of the formulas presented by Fano and Nozières are collected in Section VI-4 of this report. The relationship between the spectra excited in aluminum by electrons and by optical radiation has recently been analyzed quantitatively in some detail by H. Mendlowitz; initial results of this analysis were presented at the meeting by R. T. McGinnies.

The general theoretical reviews were followed by reviews of experimental work presented by Leder and by Watanabe. Outlines of these reviews, prepared by their authors, are given in subsequent sections of this report. Both reviews concerned the dependence of the main peaks in the spectra of energy losses upon various factors. Watanabe showed a small dependence of the peak position on the foil thickness; the reported precision in the determination of this position ( $\pm 0.1$  ev)--which was necessary to detect the effect--

was perhaps the most astonishing result presented in these sessions. Leder discussed in particular the comparison of materials with the same element in different chemical forms, the dependence on temperature, details of the dependence on the electron deflection, and a comparison between single crystals (which are anisotropic) and polycrystalline samples of the same element. Watanabe also reported on the systematic variation of the main peak with the angle of deflection and the angle of incidence of the electron on a foil.

In the last session of the meeting, Ferrell discussed the effects of the boundaries of a foil on the energy losses of electrons traversing it. These effects, first pointed out by Ritchie,<sup>120</sup> consist of: (a) small shift of the energy losses with respect to those that would occur in an infinitely thick sample of the same material, and (b) the appearance of additional levels of energy loss corresponding to excitations confined to the proximity of the foil surfaces. Detailed studies of these surface excitations are given in a paper by Ferrell,<sup>121</sup> the content of which coincides almost completely with the report presented at the meeting. In particular, Ferrell pointed out that one of the types of excitation which are produced by electrons traversing a foil is coupled to electromagnetic waves traveling outward from the foil surface and may therefore be detected optically.

The final report of the meeting, by Brandt, dealt primarily with the theoretical prediction of the energy losses of electrons in LiH. This substance is of particular interest because it is the solid insulator with the smallest number of electrons per atom (except for solid H<sub>2</sub>) and because it should have a particularly simple set of excitation levels. This discussion of LiH has already been published.<sup>122</sup> Brandt also discussed his point of view on "collective" effects in the motion of electrons within individual atoms, and on their influence on the energy losses by incident electrons. This point of view, which underlies his results on LiH and those on the stopping power constant I (see Section II - 2 of this report), has been the object of disagreement at the meeting and in subsequent correspondence. A further discussion will be found in the Appendix.

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<sup>120</sup>R. H. Ritchie, Phys. Rev. 106, 874 (1957).

<sup>121</sup>R. A. Ferrell, Phys. Rev. 111, 1214 (1958).

<sup>122</sup>W. Brandt, Phys. Rev. 111, 1042 (1958).

## 2. Measurements of Characteristic Energy Loss of Electrons

L. B. Leder, L. Marton, H. Mendlowitz,  
J. Arol Simpson, and M. D. Wagner  
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The investigation of the characteristic energy losses being pursued at the National Bureau of Standards is under the direction of Dr. L. Marton, Chief of the Electron Physics Section. We have been engaged in this problem since 1951. In addition to the authors, Dr. B. Gauthe of the University of Paris spent one year in our laboratory working on this problem.

When we first started this work we were engaged only in measuring the energy loss values for the metals and some few complex materials.<sup>123</sup> Since most of these values were not known at the time this in itself was important. However, with time many questions presented themselves, as they have a habit of doing, and the investigation expanded in an attempt to answer these questions. To this date we have only a few answers. In the following we will attempt to give a summary of what we have found. This information can be broadly outlined as follows: (a) Energy loss values. What we want to discuss is not the energy loss values themselves, but rather the effects which may or may not influence the values. (b) Angular dependence of the energy losses and of the intensities. (c) Cross sections. These are probably the least well-known of the information needed. We have, however, been able to make some relative cross section measurements. (d) Interpretation. We will not attempt to discuss theory, but only point out some relationships which we have found between the energy loss values of electrons and other types of measurements.

During the course of measuring the spectra of various materials we noted that there was a decided similarity between the spectra of some compounds and the spectra of the metals forming the compound.<sup>124</sup> This was found to be so for Sb, Ca, Pb, Te, Na, and Si.

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<sup>123</sup>L. Marton and L. B. Leder, Phys. Rev. 94, 203 (1954).

<sup>124</sup>L. B. Leder, Phys. Rev. 103, 1721 (1956).



In the case of Al and Mg there did not appear to be any similarity. In general all the features of the metal spectrum are repeated in the spectrum of the compound. The only differences are in the relative intensities (the intensities are not absolute since the spectra were measured from uncalibrated photographic plates) and an increase in the energy losses in the compound.

Measurements were also made to observe whether the characteristic loss value was affected by thickness.<sup>125</sup> For five aluminum samples varying in thickness from 35 to 500 Å no change in the energy loss value was noted. These same measurements were also used to check the suggestion of Ritchie that the 7 ev loss in Al could be a "sub-plasma" loss;<sup>126</sup> that is, a lowering of the main loss caused by the depolarization effect of the finite grain boundaries. This loss would appear at a value  $1/\sqrt{2}$  or approximately .71 times the main loss if only the thickness were finite, and  $1/\sqrt{3}$  or approximately .58 times the main loss if the grains are assumed spherical (finite in all directions). Instead it is found that the ratio of the losses is on the order of  $1/\sqrt{5}$  or .45. A further check was the variation of the ratio of the intensities of the two losses as the thickness was changed. The results did not verify the theoretically expected variation. Ritchie's calculation was made for the total cross section while our measurements were made for a solid angle of approximately  $10^{-6}$  steradian in the forward direction. However, we feel that the comparison is justified since it is experimentally known that practically all of the scattered intensity is encompassed in a solid angle of the order of  $10^{-5}$  to  $10^{-8}$  steradian at the zero position.

We have recently completed some work on measuring the temperature variation of the characteristic energy loss for Al.<sup>127</sup> In the region above room temperature this fits quite well the variation expected on the basis of a change in the density of electrons due to thermal expansion. However, in the low temperature region we found a considerable deviation from the expected value. Because of other commitments we have not as yet pursued this investigation

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<sup>125</sup>L. Marton, J. A. Simpson, J. A. Suddeth, M. D. Wagner, and H. Watanabe, Phys. Rev. 110, 1057 (1958).

<sup>126</sup>R. H. Ritchie, Phys. Rev. 106, 874 (1957).

<sup>127</sup>L. B. Leder and L. Marton, Phys. Rev. 112, 341 (1958).

further, but we believe that we can now make some predictions which we hope to test in the near future. One is that the point where the measured values begin to deviate from the simple density effect is related to the Debye temperature. Second, that for a metal such as Pb which has a Debye temperature near liquid nitrogen temperature there will be a small effect, and for a metal such as Be which has a high Debye temperature ( $1000^{\circ}$  K) the measured values will fall below the density effect values at high temperature, but that in the low temperature region the effect will also not be as great as for Al. This is because it is expected that the change with decreasing temperature will reach a maximum value and in the case of Be this should occur rather soon below liquid nitrogen temperature. It is also expected that a more detailed study of Al will show a leveling off somewhere close to liquid helium temperature or slightly below.

Angular distribution measurements: The angular distribution measurements are three dimensional, involving angle, energy, and intensity. To represent these we use a cartographic form<sup>128, 129</sup> where the ordinate is energy, the abscissa is angle and the contour lines are levels of equal intensity. In general, the intensity drops off very rapidly with angle, the intensity of the no-loss line being greater until we reach an angle of between  $2 \times 10^{-2}$  to  $9 \times 10^{-2}$  degrees at which point the loss line becomes stronger, until at the first diffraction ring the situation is again reversed. The maximum ratio of inelastic intensity to elastic intensity occurs between  $5 \times 10^{-3}$  and  $10^{-2}$  radians. This position does not appear to be a function of thickness.

It is interesting to compare the angular distribution for a non-metallic material. In the case of collodion it is found that the features are less pronounced than for the metals. The fall-off from zero angle is slower and the energy loss broader and weaker. However, a large angular dispersion is found.

We have also made measurements comparing the angular distribution for polycrystalline and single crystal metals.<sup>129</sup> In our earlier measurements this was done for gold. In more recent

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<sup>128</sup>L. Marton, J. A. Simpson, and T. F. McCraw, Phys. Rev. 99, 495 (1955).

<sup>129</sup>J. A. Simpson, T. F. McCraw, and L. Marton, Phys. Rev. 104, 64 (1956).

work<sup>125</sup> we have compared the scattering for aluminum polycrystal and single crystal, and again find a difference between the two. If we plot the ratio elastic intensity to characteristic loss intensity at zero angle and at  $6 \times 10^{-2}$  radians, which is the position of the 220 diffraction maximum, as a function of film thickness, several things can be ascertained. First, we note that the scattering intensities are different at a diffraction maximum than at zero angle. We also note that this difference depends on the thickness of the specimen, being greater for thin films and becoming small at large film thicknesses. We can also tell that the intensity ratio is more affected at zero angle than at 220 by film thickness. Finally we can say that the single crystal film shows less probability of inelastic scattering than does the polycrystalline film. It is also observed that there is a large number of inelastically scattered electrons in the diffraction maximum. This can be of considerable importance in precision electron diffraction work since it would have the effect of moving the diffraction maximum to larger angles in polycrystalline samples, and with increasingly thicker specimens. Lattice spacing calculations from such diffraction measurements could, therefore, be in error.

It was first pointed out by Cauchois that there might be a relationship between the X-ray fine structure which occurs on the high energy side of the X-ray absorption edge and the characteristic energy losses. However, she incorrectly compared the fine structure minima instead of the maxima (of absorption) with the energy losses. This was pointed out by Watanabe. We undertook a comprehensive survey of this question, and found that there was indeed a rather striking coincidence of values considering the inadequacy of the two sets of data.<sup>130</sup> There are cases where the comparison is not good. One such case is Be and another is Ca. So far it is not clear why these latter cases show this discrepancy. It can also be shown that the lattice spacing has some effect on the energy loss values.

Another set of data that are related to the characteristic energy losses are the optical absorption measurements. These have been made only for a few materials in the energy range we are interested in, the ultraviolet and far ultraviolet. In the case of aluminum, for

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<sup>130</sup>L. B. Leder, H. Mendlowitz, and L. Marton, Phys. Rev. 101, 1460 (1956).

instance, it is quite well established<sup>131</sup> that the reflectance tends to vanish at approximately 800 Å. This corresponds to the 15 ev loss. There are also some older measurements by Wood and Lukens<sup>132</sup> available for Li, Na and K which give the limits at 1550, 2100 and 3150 Å respectively. However, Wood states in the article that the limit cannot be sharply defined. A more accurate determination of the limits for Na and K can be obtained from the measurements of  $\bar{n}$  and  $\bar{\kappa}$  by Ives and Briggs.<sup>133</sup> A plot of  $\bar{n}^2 - \bar{\kappa}^2$  versus wavelength gives the values 5.6 ev for Na and 3.7 ev for K at the point where  $\bar{n}^2 - \bar{\kappa}^2$  goes to zero. These are in excellent agreement with the measured energy loss value of  $5.5 \pm .1$  ev and  $3.8 \pm .1$  ev. In the case of lithium the agreement with Wood's value is not as good. The optical value is 8.0 ev while the average loss value was found to be  $9.5 \pm .4$  ev.

It has been suggested<sup>126, 134</sup> that the loss found at 7 ev in aluminum may be due to (1) sub-plasma surface effects or (2) contamination. We have noted earlier that experiment shows that surface effects cannot account for this loss, and experiment at elevated temperatures, where contamination is reduced considerably, rules out the latter. It has been shown<sup>135</sup> that the 7 ev loss is consistent with the optical measurements of Hass, Hunter, and Tousey.<sup>131</sup> By further application of the dielectric constant model<sup>136</sup> oscillator strengths can be estimated for the "free electron" conductivity losses and optical transitions. Preliminary calculations show that the "free" conduction electrons in aluminum are less than three and closer to two per atom, in agreement with conductivity measurements. This model explains both the 7 ev and 15 ev losses in aluminum.

<sup>131</sup>Hass, Hunter, and Tousey (unpublished); Walker, Samson, and Rustgi, J. Opt. Soc. Am. 48, 71 (1958); Hass, Hunter, and Tousey, J. Opt. Soc. Am. 46, 1009 (1956).

<sup>132</sup>R. W. Wood and C. Lukens, Phys. Rev. 54, 332 (1938).

<sup>133</sup>H. E. Ives and H. B. Briggs, J. Opt. Soc. Am. 26, 238 (1936); *ibid.*, 27, 181 (1937).

<sup>134</sup>Lupton, Ferrell, and Meyers, Bull. Am. Phys. Soc. 3, 191 (1958).

<sup>135</sup>H. Mendlowitz (to be published).

<sup>136</sup>Marton, Leder, and Mendlowitz, Advances in Electronics and Electron Physics 7, 184 (1955).



### 3. Summary of Energy Loss Experiments

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Recent years have seen a rapid development in the observation and interpretation of the energy loss of electrons passing through solids. The initial energy of the electrons used is ordinarily in the region of 10 to 50 kev, and we are especially interested in energy losses of the order of 10 to 25 ev, the scattering angle ranging up to  $10^{-1}$  radian. The energy-transfer mechanism in this energy and scattering region is believed to be closely related to the plasma-oscillation theory of electrons in solids as proposed by Bohm and Pines. For this reason the experiment has attracted the attention of both theoretical and experimental physicists.

Some of the data which will be discussed in this report are as follows: (a) value of the characteristic energy loss, (b) comparison with optical data, (c) angular dependence of the characteristic loss, (d) dependence on the film thickness, (e) dependence on the incident angle.

(a) Value of the Characteristic Energy Loss. In Figure 64 the experimental results are shown in a pictorial representation. The energy measurements were carried out by analyzing the electrons scattered within an angle of  $10^{-2}$  radian. The most important point is that every substance so far examined has a characteristic energy loss of the order of 10 to 25 ev. Some of the loss lines are surprisingly sharp (nearly equal to the line width of the primary beam), but most of them are broad (more than 10 ev). The sharpness of loss lines is sometimes the focus of discussion on the origin of the energy loss concerned.

Ruthemann<sup>137</sup> examined the dependence of the characteristic loss of Al at about 15 ev upon the primary energy, and concluded that practically no dependence was observed. Quite recently

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<sup>137</sup>Ruthemann, Ann. Physik 2, 113 (1948).



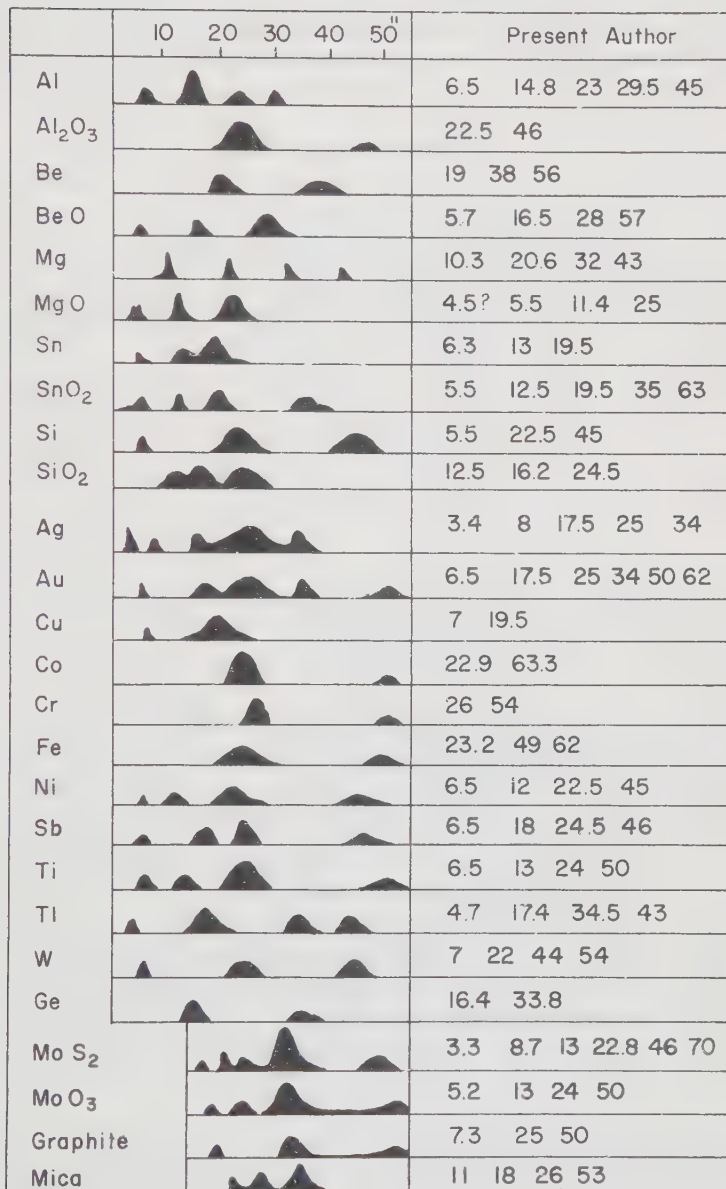


Figure 64. Characteristic energy-loss spectra.

Arai<sup>138</sup> carried out a precise measurement of the loss value and found a slight decrease with the increase of primary energy from 15 to 20 kev. His results are given in Table XVI. We have not yet a reasonable interpretation for these experimental results.

TABLE XVI

Comparison of the Energy Loss for  
Primary Energy of 15 kev and 20 kev

| Primary Energy<br>(kev) | specimen A<br>t = 140 Å<br>θ = 0° | specimen B<br>t = 420 Å<br>θ = 0° | specimen B<br>t = 420 Å<br>θ = 45° |
|-------------------------|-----------------------------------|-----------------------------------|------------------------------------|
| 15                      | 14.78±0.01<br>19*                 | 14.84±0.01<br>18*                 | 14.89±0.01<br>16*                  |
| 20                      | 14.70±0.01<br>12*                 | 14.76±0.01<br>17*                 | 14.79±0.01<br>18*                  |

\*Indicates the number of measurements.

(b) Comparison with the Optical Data. The energy loss mechanism is sometimes discussed in the framework of the theory of optical absorption and dispersion. According to H. Fröhlich and H. Pelzer<sup>139</sup> the sharp peak in the  $n / (n^2 + \kappa^2)^2$  vs  $\lambda$  curve predicts the existence of the corresponding electron energy loss. Actually in case of Ag a sharp maximum is observed in the curve at  $\lambda = 330\text{m}\mu$  ( $h\nu = 3.8$  ev), and correspondingly a fairly sharp loss line is observed at 3.5 ev in the energy-loss experiment. Most metals show the maximum in the far ultraviolet region where the optical measurement of these constants is so difficult that we have very few data at present. Optical studies in this region are strongly desired.

(c) Angular Dependence of the Energy Loss Value. Some of the characteristic energy loss values increase with the scattering angle. For the first time I observed this with a modified Mollenstedt-type electron-velocity analyzer which can analyze the

<sup>138</sup>Arai, Science Reports of Tohoku University, 41, 209 (1958).

<sup>139</sup>H. Fröhlich and H. Pelzer, Proc. Phys. Soc. A68, 525 (1955).

energy distribution of the electrons composing an electron diffraction pattern.<sup>140</sup> Dr. Marton's group<sup>141</sup> confirmed the same result with a specially designed magnetic analyzer--slowing down, analysis and reaccelerating, and Dr. Raether's group<sup>142</sup> did it with a retarding potential method.

The energy loss value  $\Delta E$  is a function of the scattering angle  $\theta$ , being represented as follows:  $\Delta E = \Delta E_0 + \underline{a} \theta^2$ . The constants  $\Delta E_0$  and  $\underline{a}$  are listed in Table XVII, in which twice the accelerating voltage is used as a unit. In the last column are listed the theoretical values for  $\underline{a}$  calculated from plasma oscillation theory. The agreement between the experimental and theoretical values is satisfactory.

TABLE XVII

Experimental Values of  $\Delta E_0$  and  $\underline{a}$  and Theoretical Value of  $\underline{a}$   
(The accelerating voltage E is 25 kev)

|          | $\Delta E_0/2E$      | $\underline{a} \ 2E$ (experimental)<br>radian <sup>-2</sup> | $\underline{a} \ 2E$ (theoretical)<br>radian <sup>-2</sup> |
|----------|----------------------|-------------------------------------------------------------|------------------------------------------------------------|
| Be       | $3.8 \times 10^{-4}$ | $0.42 \pm 0.04$                                             | 0.45                                                       |
| Mg       | $2.1 \times 10^{-4}$ | $0.62 \pm 0.04$                                             | 0.44                                                       |
| Al       | $3.0 \times 10^{-4}$ | $0.50 \pm 0.05$                                             | 0.45                                                       |
| Ge       | $3.3 \times 10^{-4}$ | $0.83 \pm 0.15$                                             | 0.45                                                       |
| Graphite | $1.5 \times 10^{-4}$ | $1.0 \pm 0.3$                                               | 0.50                                                       |

(d) Dependence of the Energy-loss Value on Film Thickness. The characteristic energy-loss value of Al at about 15 ev depends upon the film thickness, as shown in Figure 65. The energy-loss value increases with the decrease of film thickness. If we assume that the crystal size is controlled by the film thickness, this experimental result may be interpreted as follows: The plasma wave length in a finite crystal is limited by the crystal size.

<sup>140</sup>J. Phys. Soc. Japan, 11, 112 (1956).

<sup>141</sup>Phys. Rev. 104, 64 (1957).

<sup>142</sup>G. Meyer, Z. Physik 148, 61 (1957).

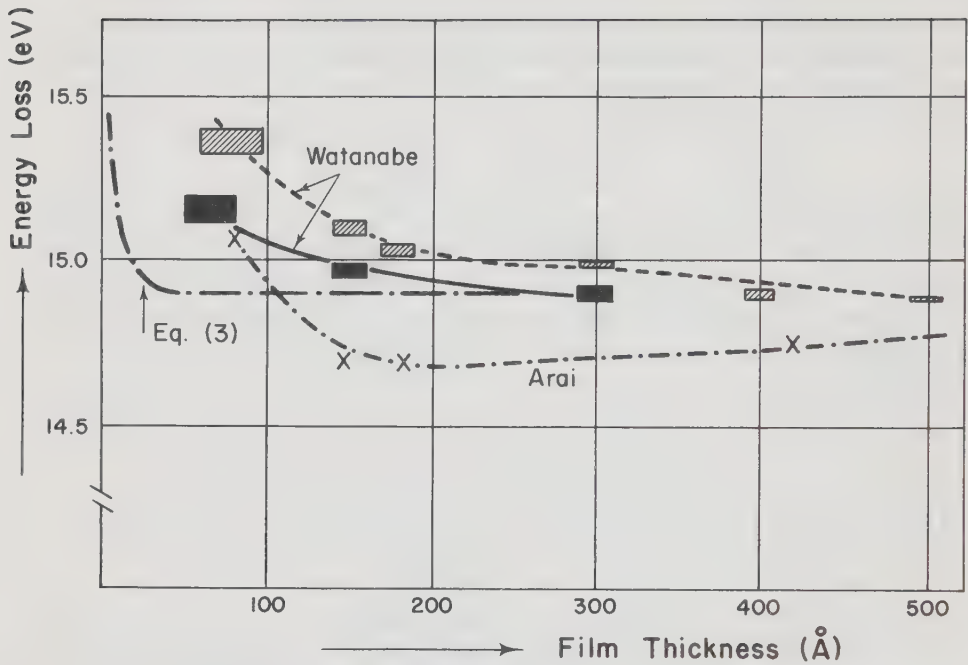


Figure 65. Dependence of energy-loss value in Al upon the film thickness.

Putting the maximum wavelength  $\lambda_{\max} = 2d$  ( $d$  is the crystal size) into the dispersion relation of the plasma wave

$$\hbar\omega = \hbar\omega_p + \frac{\langle T \rangle_{av}}{\hbar\omega_p} \frac{\hbar^2}{m} \frac{4\pi^2}{\lambda^2} \quad (2)$$

we have a relation between the plasmon energy  $\hbar\omega$  and the crystal size  $d$ ,

$$\hbar\omega = \hbar\omega_p + \frac{\langle T \rangle_{av}}{\hbar\omega_p} \frac{\hbar^2}{m} \frac{\pi^2}{d^2} \quad (3)$$

This relation is plotted in Figure 65. It shows a behavior similar to the experimental curve, but the increase occurs at smaller crystal size compared to experiment. This discrepancy may be due to the fact that the abscissa is not the crystal size itself but the film thickness, in our experiment. In other words, the crystal size may be much smaller than the film thickness.

(e) Dependence of the Energy-loss Value on the Incident Angle.

It is an interesting problem to know the dependence of the energy-loss value on the incident angle, especially in case of substances having a strongly anisotropic electronic structure. From the experimental result, we may obtain information on the anisotropy of the electronic structure.

A thin single-crystal foil of graphite was examined. The 25 kev electrons are incident on the foil at an angle  $\theta$  with the c-axis which is perpendicular to the foil. The characteristic energy loss of about 7 ev was precisely measured at different incident angles of  $\theta = 0^\circ$  and  $\theta = 45^\circ$ . The experimental results are as follows:

| <u>Incident angle</u> | <u>Energy-loss value</u> | <u>Number of measurements</u> |
|-----------------------|--------------------------|-------------------------------|
| $0^\circ$             | $6.8 \pm 0.2 \text{ ev}$ | 9                             |
| $45^\circ$            | $6.8 \pm 0.2 \text{ ev}$ | 13                            |

According to Ichikawa<sup>143</sup> the plasma frequency of  $\pi$ -electrons in graphite is given as

$$\omega_p^2 = \frac{4\pi ne^2}{m} \left[ 1 - \frac{m}{m(\theta)^*} \right] \quad (4)$$

where

$$\frac{1}{m(\theta)^*} = \frac{1}{2\hbar^2} \left[ \sin^2 \theta + \frac{1}{2} \frac{\gamma_1}{\gamma_0} \left( \frac{c}{a} \right)^2 \cos^2 \theta \right] a^2 \gamma_0 \quad (5)$$

and  $\gamma_0$ ,  $\gamma_1$  are the resonance energies between the nearest neighbors in the plane and between planes, respectively.

The  $\gamma_1$  can be estimated at 0.32 ev from the energy-loss value at  $\theta \neq 0$ , being consistent with currently estimated values of 0.2-0.5 ev. From the experimental result showing no  $\theta$  dependence, we can estimate the value of  $\gamma_0$  at about 1.6 ev. This is consistent with the value deduced from other experiments.<sup>144</sup>

This kind of discussion gives information on the electronic structure of the substance. Theoretical as well as experimental work along this line is strongly desired.

<sup>143</sup>Ichikawa, Phys. Rev. 109, 653 (1958).

<sup>144</sup>For example: J. Hove, Phys. Rev. 100, 645 (1956).



#### 4. Compilation of Dielectric Constant Formulas Presented by Nozières and Fano

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The generalized complex dielectric constant  $\epsilon(k, \omega)$  of a macroscopically homogeneous isotropic material may be introduced as was done by Nozières and Pines for an electron gas.<sup>145</sup> One requires that the electric field generated in the material by an "external" sinusoidal charge distribution with density  $e s_k \exp i(\mathbf{k} \cdot \mathbf{r} - \omega t)$  obey the Maxwell equation

$$i\epsilon(k, \omega) \mathbf{k} \cdot \mathbf{E} = 4\pi e s_k e^{i(\mathbf{k} \cdot \mathbf{r} - \omega t)} \quad (1)$$

This dielectric constant can be expressed in terms of the operator  $\rho_k = \sum_i \exp -i \mathbf{k} \cdot \mathbf{r}_i$  where  $\mathbf{r}_i$  is the position of the  $i$ -th electron of the material. More specifically, it is given in terms of the matrix of this operator in the scheme of energy eigenstates  $n$  of the unperturbed material by

$$\frac{1}{\epsilon(k, \omega)} = 1 + \frac{4\pi e^2}{\hbar k^2} \sum_n \left| (\rho_k)_{no} \right|^2 \left( \frac{1}{\omega - \omega_{no} + i\eta_n} - \frac{1}{\omega + \omega_{no} + i\eta_n} \right) \quad (2)$$

where  $\hbar \omega_{no}$  is the excitation energy of the  $n$ -th level and  $\eta_n$  its damping constant. In the limit of  $\eta_n = 0$ , (2) becomes

$$\frac{1}{\epsilon(k, \omega)} = 1 - \frac{4\pi e^2}{\hbar k^2} \sum_n \left| (\rho_k)_{no} \right|^2 \left\{ P \left( \frac{2\omega_{no}}{\omega^2 - \omega_{no}^2} \right) + i\pi [\delta(\omega - \omega_{no}) + \delta(\omega + \omega_{no})] \right\} \quad (2')$$

The connection between  $\epsilon(k, \omega)$  and the Van Hove autocorrelation function is given by reference 148, equation (9).

In the limit of large  $\omega$ ,  $\epsilon$  depends only on the density  $N$  of electrons in the material,

<sup>145</sup>P. Nozières and D. Pines, Nuovo Cimento, 9, 470 (1958).

$$\epsilon(\omega) \xrightarrow{\omega=\infty} 1 - \frac{\omega_p^2}{\omega^2}, \quad \frac{1}{\epsilon(\omega)} \xrightarrow{\omega=\infty} 1 + \frac{\omega_p^2}{\omega^2}, \quad \text{with } \frac{\omega_p^2}{\omega^2} = \frac{4\pi N e^2}{m} \quad (3)$$

From this behavior follows the sum rule

$$\int_0^\infty \text{Im}(\epsilon) d(\omega^2) = \int_0^\infty \text{Im}(-1/\epsilon) d(\omega^2) = \omega_p^2 \quad (4)$$

Approximate calculations of  $\epsilon$  for an electron gas are discussed in reference 145.

Probability of energy losses. When a particle of charge  $e$ , momentum  $p$ , and velocity  $v = \beta c$  traverses the material, the probability per unit path length of an inelastic collision with energy loss  $\hbar\omega$  and (small) deflection  $\theta$ , per unit  $d(\hbar\omega)$  and per unit solid angle  $d\Omega$  is represented by the differential coefficient

$$\frac{d^3\tau}{d(\hbar\omega)d\Omega} = \frac{e^2}{\pi^2 \hbar^2 v^2} \frac{1}{\theta_0^2 + \theta^2} \text{Im} \left\{ -\frac{1}{\epsilon(k, \omega)} + \frac{\beta^2 \theta^2}{\theta_0^2 [1 - \beta^2 \epsilon(k, \omega)] + \theta^2} \right\} = (5)$$

$$= \frac{e^2}{\pi^2 \hbar^2 v^2} \frac{\epsilon_2(k, \omega)}{\theta_0^2 + \theta^2} \left\{ \frac{1}{\epsilon_1^2 + \epsilon_2^2} + \frac{\beta^4 \theta_0^2 \theta^2}{[\theta_0^2 (1 - \beta^2 \epsilon_1) + \theta^2]^2 + \beta^4 \epsilon_2^2 \theta_0^4} \right\} \quad (5')$$

where

$$\theta_0 = \frac{\hbar\omega}{pv}, \quad k = \frac{p}{\hbar} \sqrt{\theta_0^2 + \theta^2}, \quad \epsilon_1 = \text{Re}(\epsilon), \quad \epsilon_2 = \text{Im}(\epsilon) \quad (6)$$

The second term in the braces of (5) represents the contribution of the electromagnetic (transverse) interaction<sup>146</sup> and is negligible unless  $\beta$  is very close to 1, it will be neglected in the following.

When the material is a low density gas, we may set  $\epsilon_1 \sim 1$  and  $\epsilon_2^2 \ll 1$ , so that the factor in the braces of (5') reduces to 1. The factor  $|\langle \rho_k \rangle_{\text{no}}|^2$  in (2) reduces to the square of the generalized form factor for a single molecule multiplied by the density of molecules in the gas, thereby (5') reduces to the Bethe formula for the rate of inelastic collisions.<sup>147</sup>

<sup>146</sup>U. Fano, Phys. Rev. 102, 385 (1956).

<sup>147</sup>H. A. Bethe, Handbuch der Physik, Springer, Berlin, 24/1, 493 (1933).

Irrespective of low-density approximation, if we enter (2) into (5) and disregard the second term in the braces of (5), we find

$$\frac{d^3\tau}{d(\hbar\omega)d\Omega} = \frac{4e^4}{p^2v^2} \frac{1}{(\theta_0^2 + \theta^2)^2} \sum_n \left| (\rho_{\mathbf{k}})_{no} \right|^2 \delta(\hbar\omega - \hbar\omega_0) \quad (5'')$$

This formula coincides with the Bethe formula, provided that the whole material is regarded as a single molecule and  $(\rho_{\mathbf{k}})_{no}$  as its generalized form factor.

Other properties of the material. The optical properties of a material are represented by its complex refraction index  $n + i\kappa$  (where  $\kappa$  = extinction coefficient) which is related to  $\epsilon$  by

$$n - i\kappa = \sqrt{\epsilon}, \quad \epsilon_1 = n^2 - \kappa^2, \quad \epsilon_2 = 2n\kappa \quad (7)$$

The incoherent scattering function,  $S(\mathbf{k})$ , represents the probability of excitation or ionization of an atomic system in a process where its electrons absorb a given momentum  $\hbar\mathbf{k}$ . This function is defined as

$$S(\mathbf{k}) = \sum_n \left| (\rho_{\mathbf{k}})_{no} \right|^2 = \frac{\hbar k^2}{4\pi^2 e^2} \int_0^\infty \text{Im} \left[ \frac{1}{\epsilon(\mathbf{k}\omega)} \right] d\omega \quad (8)$$

The probability of Compton scattering with deflection  $\theta$  by the atomic electrons of a material, per unit path of incident photons of frequency  $\nu$ , is approximately equal to the cross section for scattering by one free electron multiplied by  $S(\frac{2\nu \sin \frac{1}{2}\theta}{c})$ . Similarly the scattering coefficient (5) integrated over  $\omega$  (disregarding the second term in the braces) yields the probability of inelastic scattering by an angle  $\theta$

$$\frac{d^2\tau}{d\Omega^2} = \frac{4e^4}{p^2v^2} \frac{S(p\sqrt{\theta_0^2 + \theta^2}/\hbar)}{(\theta_0^2 + \theta^2)^2} \quad (9)$$

The coulomb interaction energy among electrons in the ground state is

$$\begin{aligned} E_{\text{int}} = (H_{\text{coul}})_{00} &= \sum_{\mathbf{k}} \frac{2\pi e^2}{k^2} \sum_n \left| (\rho_{\mathbf{k}})_{no} \right|^2 = \\ &= \sum_{\mathbf{k}} \frac{2\pi e^2}{k^2} \int_0^\infty \text{Im} \left[ -\frac{1}{\epsilon(\mathbf{k}\omega)} \right] d\omega \end{aligned} \quad (10)$$

This interaction energy, and  $\epsilon$  itself, may be regarded as functions of the electronic charge  $e$  if this be treated as a variable coupling constant. By so doing the total ground state energy of the material is related formally to the energy that would obtain in an independent electron approximation,

$$E_o(e^2) = E_o(o) + \int_0^{e^2} \frac{d(e'^2)}{e'^2} E_{int}(e'^2) \quad (11)$$

The characteristic energy losses in condensed matter. The main dependence on  $\hbar\omega$  of the probability of energy losses is due to the factor  $\epsilon_2/(\epsilon_1^2 + \epsilon_2^2)$  in (5'). Notice that this factor can be large only at frequencies  $\omega$  at which both  $\epsilon_1$  and  $\epsilon_2$  are small. At the same time the integral of this factor over the spectrum is fixed and equal to the integral over  $\epsilon_2$ , owing to the sum rule (4).

In the simple case of a dense electron gas (plasma model),  $\epsilon_2(\omega)$  is concentrated in a single peak at  $\omega = 0$ , and  $\epsilon_1$  is large and negative at small  $\omega$  and vanishes at  $\omega = \omega_p$  ("plasma frequency") so that  $\epsilon_2/(\epsilon_1^2 + \epsilon_2^2)$  also peaks there. In most of the actual materials,  $\epsilon_2$  has not just one but a series of peaks or broad maxima in the optical range, corresponding to intra- and inter-band transitions. It follows that the spectrum of energy losses, represented by  $\epsilon_2/(\epsilon_1^2 + \epsilon_2^2)$  should also show a series of peaks in this range. However the following considerations indicate that these peaks must be small.

Owing to the dispersion relations between the real and imaginary parts of  $\epsilon$ , i. e., between  $\epsilon_1$  and  $\epsilon_2$ , it can be shown that average values  $\langle \epsilon_2/(\epsilon_1^2 + \epsilon_2^2) \rangle$  obtained by smoothing out a number of adjacent peaks are related approximately to corresponding averages of  $\epsilon_1$  and  $\epsilon_2$  by

$$\left\langle \frac{\epsilon_2}{\epsilon_1^2 + \epsilon_2^2} \right\rangle \sim \frac{\langle \epsilon_2 \rangle}{\langle \epsilon_1^2 \rangle + \langle \epsilon_2^2 \rangle} \quad (12)$$

That is, the mutual exclusion between large values of  $\epsilon_2$  and  $\epsilon_2/(\epsilon_1^2 + \epsilon_2^2)$  extends also to their average values. Now, the value of  $\epsilon_2$  averaged over the whole optical range, say from  $\omega = 0$  to  $\omega = R$ , where  $R$  is the Rydberg frequency ( $\hbar R = 13.6$  ev), is seen to be larger than 1 for practically all condensed materials. This follows from the fact that the fraction of the  $\int_0^\infty \epsilon_2 d(\omega^2)$  in (4) which is contributed by the integration over the optical range should equal approximately the fraction of the factor  $N$  in  $\omega^{-2}$  which is contributed by the valence electrons in the material. We set accordingly

$$\langle \epsilon_2 \rangle = \frac{1}{R^2} \int_0^{R^2} \epsilon_2 d(\omega^2) \sim \omega_p^2 \frac{Z_{\text{val}}}{Z} = \frac{4\pi e^2}{m} \frac{N}{Z} Z_{\text{val}} = 14 \frac{\rho}{A} Z_{\text{val}} \quad (13)$$

where  $\rho$  and  $A$  are the density (in  $\text{g/cm}^3$ ) and the atomic weight of the material. For most condensed materials we have  $14\rho/A \sim 2$  and  $Z_{\text{val}} \geq 1$ . Therefore  $\epsilon_2/(\epsilon_1^2 + \epsilon_2^2)$  can have no major peak in the optical range. The characteristic energy loss, which corresponds to the contribution of optical electrons to the

$$\int_0^\infty [\epsilon_2/(\epsilon_1^2 + \epsilon_2^2)] d(\omega^2)$$

must therefore occur at a somewhat higher energy, just above the optical range, where both  $\langle \epsilon_2^2 \rangle$  and  $\langle \epsilon_1^2 \rangle$  have become small. This energy will vary depending on the number of valence electrons.



## APPENDIX\*

### Comments on "Collective" Effects in Atoms and in Extended Media

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The study of "plasma-type" collective effects of interaction among electrons in extended media has led some workers, notably Lindhard and Brandt among the participants at the conference, to consider the possible occurrence of analogous effects in isolated atoms. The question then arises whether or not such effects are already implicitly taken into account in the Bethe theory, i. e., whether this theory is complete within the stated limits of the Born approximation.

More specifically, the question may be stated as follows. The Bethe theory defines the constant  $I$  of the stopping-power formula for a low-density monatomic gas as:

$$I = \prod_n^{\text{all } n} (\hbar \omega_{n0})^f n_0 \quad (1)$$

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\*As indicated on page 146, a disagreement emerged at the conference regarding the role of collective effects in the stopping power of atoms. Subsequent to the conference and in connection with the preparation of these Proceedings, an effort was undertaken through private correspondence to clarify the situation and to narrow down the disagreement. Following a preliminary circulation of this Appendix, it appears that very little, if any, matter of substance remains unresolved. Further clarification of this matter may, of course, be provided by additional investigations. This Appendix is accordingly presented for the information of the reader, but should not be regarded as a part of these Proceedings, having been prepared as an informal aftermath to the conference.

where  $\omega_{n0}$  indicates the frequency of the optical (or x-ray) transition from the ground state to an excited (or ionized) state  $n$  and  $f_{n0}$  is the corresponding oscillator strength. Thereby  $I$  is determined in terms of optical constants (the fact that these are poorly known theoretically or experimentally is not relevant to the question at hand). The opinion has been expressed by Brandt (in private correspondence) that the theory leading to (1) is incomplete, in that it is based only on an independent-particle model of the atom and does not specify that the states excited by charged particles are (a) stationary and (b) identical with the optical levels.

Re-examination of Bethe's article<sup>147</sup> and of the related article by Wentzel which formulates the Born theory of collisions<sup>148</sup> shows that the states "n" are indeed stated to be stationary and to be defined as solutions of the complete Schroedinger equation for a  $Z$ -electron atom (including, of course, the interaction between electron pairs). Such stationary states can be reached from the ground state under the influence of interaction with electromagnetic or corpuscular radiation. This reviewer concludes that (1) is, in fact, free from the limitations surmised by Brandt.

Even though the usually accepted interpretation of the Bethe formula is reaffirmed, it may be of interest to consider how one is led to attempt to distinguish contributions to  $I$  from different mechanisms. Consider first a degenerate electron gas (i. e., the plasma model of metal electrons). The excitations produced in this system by a fast electron may be regarded as harmonic oscillations of a polarization wave, in which the restoring force is provided by the collective effect of coulomb interactions among distant electrons. These oscillations are longitudinal, in that the electric polarization of the material is parallel to its own gradient (i. e., to the wave vector). Optical excitations of the same system are, on the contrary, transverse and are unaffected by long-range coulomb interactions, so that their spectrum begins at zero frequency.

When the binding of electrons within atoms is taken into account, a longitudinal polarization wave involves a restoring force that consists of two components: one due to coulomb interaction of distant electrons and one due to intra-atomic forces. Only the intra-atomic forces are involved in optical transitions, which may now be intra- or inter-band. In the theory of harmonic oscillations, different restoring forces contribute additively to the squared

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<sup>148</sup>G. Wentzel, Handbuch der Physik, Springer, Berlin, 24/1, 714 ff (1933).

oscillation frequency. Thus one is led to regard the squared frequency of the plasma-type excitations produced by fast electrons in an insulator as the sum of two terms; one of these equals the squared frequency of an interband (optical) transition and represents the influence of intra-atomic forces, the remainder represents the influence of long-range coulomb interactions (collective effect).

Interestingly enough, one arrives at the same result starting from an opposite point of view which regards a condensed material as a gas of separate atoms at a rather high density. In the limit of low density, incident electrons and optical radiation would excite the atoms to the same levels which have been indicated above by  $\hbar \omega_{n0}$ . As the density increases, the coulomb interaction among atoms--even distant ones--becomes important and influences the energy level of longitudinal excitations, namely of those which are produced by incident electrons and in which the atomic electrons are polarized in the direction of the momentum transfer. The resulting energy level  $\hbar \Omega_{n0}$  is related to the corresponding energy level of the isolated atoms, to first order in the atom density  $N$ , by reference 149.

$$\Omega_{n0}^2 = \omega_{n0}^2 + \frac{4\pi N e^2}{m} f_{n0} \quad (2)$$

Here again, the squared frequency is represented as the sum of two terms, one due to intra-atomic effects and the other to collective effects.

It appears, then, attractive to seek a general procedure for splitting the squared frequency corresponding to characteristic energy losses into a sum of terms corresponding respectively to intra-atomic and collective effects. However, the approach to this analysis from the plasma model, in the form indicated above, is wholly qualitative, and the opposite approach through equation (2) is valid only to first order in  $N$  (in fact, even the correspondence between the levels  $\hbar \Omega_{n0}$  and  $\hbar \omega_{n0}$  exists only in this order). Notice also that some arbitrariness is involved in the model leading to (2), in that, e.g., the interaction between electrons of the same atoms is included in the determination of  $\omega_{n0}^2$  and the interaction between electrons of different atoms is included in the other term. A straightforward treatment of electron collisions in accordance with the Bethe theory, but applied to a block of material rather than to a single atom, leads directly to (5'') on page 160 in which the

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<sup>149</sup>U. Fano, Phys. Rev. 103, 1202 (1956).

frequency  $\omega_{n0}$  is the same as that which we have called here  $\Omega_{n0}$ . The states  $n$  of that equation are states of longitudinal excitation and are attained from the ground state owing to the perturbation  $e_k = \sum_i \exp(-ik \cdot r_i)$  where  $\hbar k = p(\theta_0^2 + \theta^2)^{1/2}$  indicates the momentum transfer in the collision. These states differ from those of optical (transverse) excitation in character and energy, a difference that disappears for atoms or small molecules where the exponentials in  $e_k$  are adequately represented by the dipole approximation  $1 - ik \cdot r$ .<sup>150</sup>

An attempt to extend the plasma theory to take into account intra-atomic forces and to apply to all atomic electrons (not only to conduction ones) was made by Lindhard and Scharff in 1953.<sup>151</sup> A difficulty was encountered at the outset, namely that the plasma theory assumes that the average electron density is nearly constant over a wavelength of its oscillations; this condition is not verified in the interior of atoms. Lindhard and Scharff proceeded nevertheless, and assumed that each portion of the electron distribution can experience oscillations in which the restoring force consists of two comparable terms. One of these terms is proportional to the local electron density in the portion of atom under consideration and represents collective effects of electron interaction. The other one is proportional to the squared orbital frequency of a single electron in that portion of an atom. These two effects were not clearly distinguished. On the contrary it seems plausible to this reviewer that the same physical factors are, in effect, being taken into account twice. On the one hand the electron density is high in the interior of atoms just because of the nuclear attraction, and on the other hand the orbital frequency of each electron in an independent particle model is adjusted, through screening parameters, so as to take approximate account of the repulsion by other electrons.

Lindhard referred, in the course of private correspondence, to the treatment of atomic oscillation frequencies carried out by Jensen<sup>152</sup> using a Thomas-Fermi model and leading to a hydrodynamic equation for the atom. Different terms of this equation

<sup>150</sup>The values of  $k$  relevant to the calculation of  $I$  are always much smaller than the reciprocal size of atomic orbitals.

<sup>151</sup>J. Lindhard and M. Scharff, Dan. Mat. Fys. Medd. 27, No. 15 (1953), Sect. 4.

<sup>152</sup>H. Jensen, Z. Physik, 106, 620 (1937).



yield separate contributions to the restoring force. However, it is notoriously difficult to assign a clear and unique physical meaning to each of these terms. And it is not clear at all that any of the effects included in a crude model of the Jensen type is disregarded in the derivation of the Bethe formula (1).

Thus, no meaningful way seems to be at hand to distinguish how much "collective" and non-collective effects contribute to the frequencies excited in atoms by electron collisions. (In an extended material the separation of the levels excited by electron impact and light absorption may serve as an index of collective effects; this requires, however, that a one-to-one correspondence is preserved between levels of the two sets, which may be possible only in special cases.) True enough, one may conceive of a calculation of the parameters  $\omega_{n0}$  and  $f_{n0}$  to be entered in the Bethe formula (1) which starts from an independent-electron approximation, i. e., from an approximation that takes the electron interactions partially into account but disregards correlations among the positions of different electrons. These correlations are known to have little effect on some atomic properties, and an appreciable effect on others. They constitute collective effects in the sense that any collective effect certainly involves a correlation between particles. Their effect on I does not seem to have been estimated; it would depend, in magnitude, on the details of the independent-particle model which is taken as an initial approximation.

As mentioned above, an extension of the Bethe theory leads to the formula (5'') on page 160, which applies to condensed matter as well as to single atoms. Its validity is restricted only by the approximations  $Ze^2/hv \ll 1$  (where  $Ze$  and  $v$  are the charge and speed of the incident particle) and  $v$  much larger than the speed of the atomic electrons. In this formula the collective effects influence only the value of the generalized form factor, which is a property of the material, and do not influence the factor that represents the interaction of the incident particle with the atomic electrons. This result agrees with Bohr's initial picture of the collisions of fast charged particles as impulsive events. The difficulty in the application of the extended and of the original Bethe formula lies in the actual evaluation of the relevant matrix element (generalized form factor) for many-electron systems. Because of this practical difficulty the Bethe theory seems to have been discounted, so that its conceptual completeness was lost sight of and that it was mistakenly classified as a one-electron approximation. This interpretation is suggested not only by the correspondence mentioned



above but also by the Lindhard essay in the volume dedicated to Bohr.<sup>153</sup> Note also in connection with possible alternatives to the Bethe theory, that the original application of the Thomas-Fermi model by Bloch aimed at evaluating the constant  $I$  through equation (1) rather than at replacing this equation.

A word might be added on the connection between the time-dependent and time-independent descriptions of the collision processes. The effect of dielectric polarization on the stopping power was first treated by Fermi through a time-dependent procedure. The very concept of screening of the charge of an incident electron by dielectric polarization evokes a time-dependent picture and also implies a collective displacement of all electrons. This point of view applies to a single atom as well as to extended matter, aside from questions of quantitative importance. On the other hand, the Bethe theory utilizes a time-independent procedure. Time-dependent and time-independent methods are wholly equivalent in quantum mechanics. Any given physical factor can be taken into account equally well by either procedure, but it may not always be easy to recognize where it appears in the two methods. This difficulty of form might be responsible for some confusion.

In conclusion, this reviewer believes that the section on collective and independent particle descriptions<sup>151</sup> of the Lindhard-Scharff paper and various statements of the Lindhard review<sup>153</sup> should be interpreted with great reservations. Specifically, any expression of the stopping power in terms of the density distribution of the atomic electrons constitutes, thus far, only a conjecture. The entire series of recent papers by Brandt<sup>154</sup> rests on a development of the Lindhard and Scharff work and apparently also on a misunderstanding of the Bethe theory; it should accordingly be reconsidered in its entirety.

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<sup>153</sup>W. Pauli, ed., *Niels Bohr and the Development of Physics*, McGraw Hill, New York, (1955), p. 185 ff. See, in particular, the statements p. 188, 3d par., p. 189, 3d par., p. 190, 2d par.

<sup>154</sup>W. Brandt, *Phys. Rev.* 111, 1042 and 112, 1642 (1958, and paper in Sect. II-2 of this report.

### Other Nuclear Science Publications

Reports and preliminary reports issued by direction of the Committee on Nuclear Science and available from the Publications Office of the National Academy of Sciences-National Research Council. Distribution of free reports limited to investigators active in this field.

No. 2. Nuclear Electric Quadrupole Moments and Quadrupole Couplings in Molecules, by B. T. Feld. 1949; 25 p., paper; free.

No. 3. Neutrons from Alpha Emitters, by H. L. Anderson. 1948; 20 p., paper; free.

No. 4. Monoenergetic Neutrons from Charged Particle Reactions, by A. O. Hanson and R. F. Taschek. 1949; 27 p., paper; free.

No. 5. Energy Levels of Light Nuclei, by T. Lauritsen. 1949; 52 p., paper; free.

No. 6. Photo-Neutron Sources, by A. Wattenberg. 1949; 16 p., paper; free.

No. 9. Relative Isotopic Abundances of the Elements, by K. T. Bainbridge and A. O. Nier. 1950; 59 p., paper; free.

No. 10. Recent Applications of Electron-Multiplier Tubes, by J. S. Allen. 1950; 14 p., paper; free.

No. 11. Physical, Biological, and Administrative Problems Associated with the Transportation of Radioactive Substances, by Robley D. Evans.

No. 12. Techniques of Particle Energy Control with Van de Graaff Accelerators, by Carl L. Bailey and John H. Williams, 1952; 8 p., paper; free. Pub. 207.

No. 13. Status Report on Standardization of Radionuclides in the United States, by George G. Manov. 1953; 20 p., paper; free. Pub. 270.

No. 14. A Handbook on Mass Spectroscopy, by Mark G. Inghram and Richard J. Hayden.

No. 15. Basic Mechanisms in Radiobiology: Physical and Chemical Aspects. 1954; 145 p., paper; \$1.50. Pub. 305. Proceedings of an informal conference of physicists, chemists, and biologists held at Highland Park, Illinois, May 7-9, 1953, to consider the nature of the physical mechanisms involved in the interaction of radiation and biological systems. There are sections on energy transfer from radiation to matter, mechanisms of energy degradation and chemical change and effects of secondary electrons, effects of electronic excitation, and the importance of radiation chemical effects in radiobiology (summary). The list of references to individual sections is collected at the end of the papers and deals mainly with work published during and since 1950.

No. 16. Mass Differences. 1954; 145 p., paper; \$2.25. Pub. 336. A reprint of five articles from the Reviews of Modern Physics (Volume 26, No. 4, 1954): "Table of Total Beta-Disintegration Energies," R. W. King; "Nuclear Disintegration Energies," D. M. Van Patter and Ward Whaling, a systematic review of currently available experimental data; "Determination of Atomic Masses by Microwave Spectroscopy," S. Geschwind, G. R. Gunther-Mohr, and C. H. Townes, presenting theory, brief discussion of experimental techniques and tabulations and a discussion of mass measurements; "Table of Alpha-Disintegration Energies of the Heavy Elements," Frank Asaro and I. Perlman, listing total alpha-decay energies with indication of means used for determinations; and "Mass Spectroscopic Atomic Mass Differences," H. E. Duckworth, B. G. Hogg, and E. M. Pennington, tables of data obtained by the doublet method.

No. 17. Basic Mechanisms in Radiobiology: Biochemical Aspects. 1955; 158 p., paper; \$p.50. Pub. 367. Proceedings of an informal conference held at Highland Park, Illinois, May 1954, to elucidate basic biochemical phenomena in radiobiological actions. Sections on the direct effect of radiations on molecules of biological importance; cellular biochemistry; enzyme and related effects in the intact cell; and changes in nucleic acid metabolism as a result of radiation.

No. 18. Basic Mechanisms in Radiobiology: Cellular Aspects. 1956; 190 p., paper; \$1.50. Pub. 450. Proceedings of an informal conference held at Bear Mountain, New York, May 1955, concerned with cellular changes, cell division, and chromosome aberrations

following irradiation; the use of radiation and other agents for a study of the mechanism of mutation production; and the relation of the morphology of the cell to its functional behavior following irradiation. References.

No. 19. Nuclear Processes in Geological Settings II. Co-sponsored with the National Science Foundation. 1956; 205 p., paper; \$2.00. Pub. 400. Summaries of papers presented at a conference held at Pennsylvania State University in September 1955. Subjects include natural variation in isotopes of H, He, O, C, S, Pb and their geologic significance, the evaluation of mineral age measurements made from the decay of U-Pb, Th-Pb, Rb-Sr and K-A, induced radioactivity and determination of the half-lives of  $\text{Th}^{232}$  and  $\text{Rb}^{87}$ . Pertinent discussion of the papers are presented. Vol. I. 1954 Conference. See below.

No. 20. Multichannel Pulse Height Analyzers, by H. W. Koch and R. W. Johnston. 1957; 205 p., paper; \$2.00. Pub. 467. Proceedings of an informal conference held September 1956 at Gatlinburg, Tennessee, to discuss multichannel pulse height analyzers and related topics. Papers and discussions cover pulse shaping and amplifiers, quantizers, storage, data presentation, and areas of future research.

No. 21. Basic Mechanisms in Radiobiology: Mammalian Aspects. 1957; 203 p., paper; \$2.00. Pub. 513. An edited transcript of an informal conference held at Highland Park, Illinois, in May 1956. The topics chosen were those considered by a panel of experts to be most representative of current interests and developments in mammalian radiobiology. The complex of interrelationships are apparent in the numerous cross connections between the five sections. Each session started with a formal lecture and evolved into a round-table discussion among the twenty-seven selected participants. Session topics and principal essayists were: Survey of the problem, by Howard J. Curtis; Radiosensitive cell systems, by Henry Quastler; Carcinogenesis, by Henry Kaplan; Life-span studies, by Hardin Jones; and Mechanisms of action, by Harvey M. Patt. Illus.; references.

No. 22. Research in Radiology, by Henry S. Kaplan. 1958; 208 p., paper; \$1.75. Pub. 571. Proceedings of an informal conference held at Highland Park, Illinois, in May 1957. This conference was a continuation of the "Basic Mechanisms in Radiobiology Series," and represented an attempt to bridge the gap between experimental radiobiology and clinical radiotherapy. Topics considered at the



conference included Factors Determining Radiosensitivity in the Cell; Factors in the Host and Tumor Bed; Factors Related to the Agent and Mode of Treatment; Possibilities of a National Experiment to Evaluate Supervoltage Therapy.

No. 23. Cosmological and Geological Implications of Isotope Ratio Variations. 1958; 187 p., paper, \$1.75. Pub. 572. Proceedings of an informal conference held at Massachusetts Institute of Technology in June 1957.

No. 24. Measurements and Standards of Radioactivity. 1958; 156 p., paper; \$1.75. Pub. 573. Proceedings of an informal conference held at Easton, Maryland, in October 1957.

No. 25. Mass Differences II. 1959; 84 p., paper; \$1.50. Pub. 649. Reprinted from Reviews of Modern Physics (Volume 29, Number 4 and Volume 30, Number 2, Part 1). A compilation of experimental atomic mass differences found from Beta Decay, Reaction Energies, Microwave Data, Alpha Decay, and Mass Doublets.

No. 26. Sector-Focused Cyclotrons, by F. T. Howard. 1960; 310 p., paper; \$2.50; Pub. 656. Proceedings of an informal research conference on the design, construction, testing and operation of sector-focused cyclotrons. Reports include discussion of ion orbit theories, calculation of complex magnetic field configuration, and factors affecting beam quality and control.

No. 27. Source Material for Radiochemistry. 1959; 23 p., paper; free. Pub. 654. This compilation is an attempt to list current source material of interest to the radiochemist. It has been compiled from lists submitted by each member of the Subcommittee on Radiochemistry. Emphasis has been placed on documents and reports of a review nature which have proven useful to members of this Subcommittee. No attempt has been made to include standard analytical reference works which of course are indispensable in planning new procedures.

No. 28. Reactors for University Research. 33 p., paper; \$0.75. Papers presented by T. J. Thompson, Robert G. Cochran, Clifford K. Beck and Edson G. Case, and H. J. Gomberg at the ASEE Annual Meeting, Berkeley, June 1958.

No. 29. Penetration of Charged Particles in Matter, by Edwin A. Uehling. 1960; 174 p., paper; \$2.00; Pub. 752. Proceedings of an informal conference held in Gatlinburg, Tennessee, in September 1958.



Research and Training Facilities for Radiobiology and for Use of Isotopes in Biology and Medicine in the United States and Canada, by D. Harold Copp. 1950; 151 p., paper; free. Courses, facilities, personnel, and research projects in progress using radioactive isotopes. The survey includes 140 universities, hospitals, and other research organizations.

Nuclear Processes in Geologic Settings I. Co-sponsored by the University of Chicago, National Research Council and the National Science Foundation. 1954; 82 p., cloth; free. Summaries of papers presented at a conference held at Williams Bay, Wisconsin in September 1953. Subjects include the decay constants of  $\text{Rb}^{87}$  and  $\text{K}^{40}$ , spontaneous fission, manifestations of a "natural neutron flux," variations in isotopic abundance of Pb, H, Si, S, Sr, Li, Br and C and their geologic and cosmological significance. Pertinent discussion of the papers is included. Vol. II. 1955 Conference. See No. 19, Nuclear Science Series above.

Nuclear Data Sheets. Issued irregularly in sets, about 150 sheets per set or 1200 sheets per year. Subscriptions accepted for calendar year only. Each sheet presents a cumulation, for a particular nucleus, of data on radioactivity, energy levels, Q values, moments, etc. The style is similar to that used in Nuclear Level Schemes (TID-5300). As additional experimental results are published replacement sheets will be issued. New data are marked with arrows. The sheets, 8 1/2 x 11 inches in size, take the place of the noncumulative 3 x 5 inch Nuclear Data Cards which were published from 1954 through 1957. The sheets are available in paper of two weights: a light-weight stock for use in binders, and a heavy-weight stock for drawer filing. Annual subscription prices: U. S. A. (parcel post) paper stock \$17, card stock \$20; elsewhere (printed-matter mail) paper stock \$17, card stock \$20; elsewhere (first-class mail) paper stock \$25, card stock \$34.

Nuclear Theory Index Cards. Issued during 1958 and 1959 in 6 sets, about 120 cards per set or 720 cards per year. Subscriptions by calendar year only. The cards issued in 1958 covered theoretical papers appearing in both 1957 and 1958. Each card (3 x 5 inches in size) contains the title of an article, authors' names, institutional affiliation, number of figures and of tables in the paper, and literature reference. Prominent code numbers show the division and subdivision under which the article has been classified. Main topics are Nuclear Structure, Nuclear Reactions, Electromagnetic Radiation, Beta Decay and Parity, Alpha Decay, and Tables and Computational Aids. Most of these topics have

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